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Historical Doses from Tritiated Water and Tritiated Hydrogen Gas Released to the Atmosphere from Lawrence Livermore National Laboratory (LLNL). Part 6. Summary Report

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Part 6. Summary Report

S. Ring Peterson

ABSTRACT

Throughout fifty-three years of operations, an estimated 792,000 Ci (29,300 TBq) of tritium have been released to the atmosphere at the Livermore site of Lawrence Livermore National Laboratory (LLNL); about 75% was tritium gas (HT) primarily from the accidental releases of 1965 and 1970. Routine emissions contributed slightly more than 100,000 Ci (3,700 TBq) HT and about 75,000 Ci (2,800 TBq) tritiated water vapor (HTO) to the total. A Tritium Dose Reconstruction was undertaken to estimate both the annual doses to the public for each year of LLNL operations and the doses from the few accidental releases. Some of the dose calculations were new, and the others could be compared with those calculated by LLNL. Annual doses (means and 95% confidence intervals) to the potentially most exposed member of the public were calculated for all years using the same model and the same assumptions. Predicted tritium concentrations in air were compared with observed mean annual concentrations at one location from 1973 onwards. Doses predicted from annual emissions were compared with those reported in the past by LLNL. The highest annual mean dose predicted from routine emissions was 34 μSv (3.4 mrem) in 1957; its upper confidence limit, based on very conservative assumptions about the speciation of the release, was 370 μSv (37 mrem). The upper confidence limits for most annual doses were well below the current regulatory limit of 100 μSv (10 mrem) for dose to the public from release to the atmosphere; the few doses that exceeded this were well below the regulatory limits of the time. Lacking the hourly meteorological data needed to calculate doses from historical accidental releases, ingestion/inhalation dose ratios were derived from a time-dependent accident consequence model that accounts for the complex behavior of tritium in the environment. Ratios were modified to account for only those foods growing at the time of the releases. The highest dose from an accidental release was calculated for a release of about 1,500 Ci HTO that occurred in October 1954. The likely dose for this release was probably less than 360 μSv (36 mrem), but, because of many unknowns (e.g., release-specific meteorological and accidental conditions) and conservative assumptions, the uncertainty was very high. As a result, the upper confidence limit on the predictions, considered a dose that could not have been exceeded, was estimated to be 2 mSv (200 mrem). The next highest dose, from the 1970 accidental release of about 290,000 Ci (10,700 TBq) HT when wind speed and wind direction were known, was one-third as great. Doses from LLNL accidental releases were well below regulatory reporting limits. All doses, from both routine and accidental releases, were far below the level (3.6 mSv [360 mrem] per year) at which adverse health effects have been documented in the literature.

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INTRODUCTION

Tritium has been released to the atmosphere from facilities at the Livermore site of the Lawrence Livermore National Laboratory (LLNL) as part of routine operations since 1953¹. Since 1973, LLNL has calculated doses from these routine emissions to the hypothetical maximally exposed individual (MEI²) or the hypothetical site-wide maximally exposed individual (SW-MEI³) and reported them in the annual environmental reports (SAERs⁴). LLNL⁵ has also had a few accidental releases of tritium to the atmosphere. A dose to a member of the public after an accidental release in 1970 was calculated and reported in the open literature (Myers et al. 1973); doses from other accidental releases were estimated. LLNL also reported measured concentrations of tritium in air and vegetation after the 1970 release in the 1971 SAER (Gudiksen 1971). Prior to reporting doses⁶, LLNL reported quarterly tritium releases⁷, both routine and accidental from the Tritium Facility and other facilities, to the Atomic Energy Commission (AEC), the Energy Research and Development Administration (ERDA), and the Department of Energy (DOE), as the parent organization was named throughout the years.

There were several reasons to conduct a comprehensive analysis of operational emissions of tritium and to calculate (and recalculate) doses to the public.

1. Doses to the public from routine emissions from LLNL were not calculated prior to 1973.
2. Routine doses to the public during and after 1974 were calculated using three different dispersion models with different assumptions.
3. Inhalation/skin absorption⁸ and ingestion dose to the public after an accidental release were only calculated explicitly for the 1970 release (Myers et al. 1973; ATSDR 2003). Inhalation dose alone was calculated after the 1985 (Howe 1985b), 1989⁹ (Brown 1989) and 1991⁹ (Galles 1991) accidental releases.
4. The dose consequences of a release of tritiated hydrogen gas (HT¹⁰) are much lower than those of an equivalent release of tritiated water vapor (HTO¹¹) when

¹ Tritium was also released to the atmosphere from the Livermore site of Sandia National Laboratories/California (SNL/CA) between 1979 and 1995 (Peterson 2007a, 2007b).

² The maximally exposed individual (MEI) is defined as the hypothetical member of the public at a fixed location who receives the greatest dose from a single source of radionuclide releases to the air.

³ The site-wide maximally exposed individual (SW-MEI) is defined as the hypothetical member of the public at a single residence, school, business, church, or other such facility who receives the greatest dose from the combination of all evaluated radionuclide source emissions, as determined by modeling.

⁴ The environmental report published annually by LLNL since 1972 will be referred to in this report by the acronym by which it is known at LLNL (SAER) that stands for “site annual environmental report”.

⁵ and SNL/CA (Peterson 2007d)

⁶ (and in addition to reporting doses once reporting began)

⁷ In spite of an exhaustive search, the records of releases that were found only dated back to mid-1956, but there is no reason to think that the reports were not issued during the first few years of LLNL operations.

⁸ Throughout this document, “inhalation” will be used to mean both inhalation and skin absorption.

⁹ This was not one of the accidental releases modeled in this tritium dose reconstruction because it was small enough relative to the quantity of tritium released routinely in this year to be modeled as part of the routine releases.

¹⁰ Throughout the report, gaseous tritium in any form (e.g., T₂, DT, and HT) will be referred to as HT.

¹¹ Throughout the report, aqueous tritium in any form (e.g., T₂O, DTO and HTO) will be referred to as HTO.

- modeled appropriately¹². Based on scientific judgment and regulatory requirements at the time the doses were calculated and reported, LLNL either did not calculate dose from routine HT releases (because inhalation dose consequences of an HT release were considered 1/25,000th of an equivalent release of HTO) or modeled HT as HTO¹³. The first approach slightly underestimates dose from a release of HT; the second approach greatly overestimates dose from a release of HT. Recently developed tritium dose models for routine releases, such as DCART (Peterson 2006), account for conversion of HT to HTO in the environment and hence can estimate dose from releases of HT as well as HTO.
5. Recently developed tritium dose models for routine releases, again such as DCART, account for dose from ingesting organically bound tritium (OBT), while regulatory models only model dose from inhalation and ingestion of HTO. Dose from ingesting 1 Bq of OBT is about 2.4 (or more) times that from ingesting 1 Bq of HTO, so to ignore dose from ingesting OBT may result in the dose being underestimated.
 6. Most accident consequence models account for dose from inhalation only. Because the ingestion pathway to dose is more important after an accidental release (especially for HT) than in an equilibrium situation, a dose model, such as UFOTRI (Raskob 1990, 1993) that accounts for tritium-specific pathways leading to ingestion dose, is required.
 7. In the past, only deterministic (i.e., a single “best estimate” value) doses were calculated. In such an analysis, the uncertainty associated with the dose is not assessed, which means that it is not possible to specify the confidence that can be placed in the prediction. On the other hand, a probabilistic analysis generates a distribution of doses from which meaningful statements can be made regarding the probability that the true dose will fall within a certain specified interval. For a probabilistic set of dose predictions with a 95% confidence interval, the most likely dose will be the mean of the distribution, but the true dose will lie between the 2.5% and 97.5% confidence limits; there will be only a 2.5% probability that the true dose will exceed the upper confidence limit¹⁴.
 8. For any assessment, because different assumptions and interpretations of the data are often made, much greater confidence can be placed in the results if two (or more) sets of predictions agree (Peterson et al. 1996; Thiessen et al. 1997).

The effort to assemble all the available background information needed to calculate annual doses for each year of LLNL operations and to calculate doses for accidental releases began in 1998 and culminates with this report.

¹² When calculating dose from released HT, the conversion of HT to HTO in the soil and subsequent emission of HTO to the atmosphere must be taken into account.

¹³ The models used by LLNL for dose predictions until 1986 only accounted for dose from inhalation. The compliance model used from 1986 onwards (Moore et al. 1979; Parks 1992) only accounted for releases of HTO and dose from inhalation and ingestion of HTO.

¹⁴ In this TDR, because of various conservative assumptions, the 97.5% confidence limit on the predicted dose is viewed as the dose that could not have been exceeded.

The intent of this Tritium Dose Reconstruction (TDR) of the Livermore site was to complete and supplement the LLNL dose history by

1. Developing an equilibrium probabilistic dose model that incorporated the latest knowledge about the environmental transfer of tritium and accounted for the conversion of HT to HTO in the environment and for dose from ingestion of OBT.
2. Compiling a complete history for the Livermore site of the routine and accidental tritium releases from all facilities that could have contributed to dose to the public.
3. Compiling a record of all the information available that would indicate the accuracy of the reported release rate estimates. This included such things as calibration data for stack flows, analytical counting data, and stack heights and diameters.
4. Estimating the fraction of each tritium release that was HT or HTO and estimating the uncertainty associated with tritium release rates from all facilities.
5. Estimating the dilution factors¹⁵ and their uncertainties for each facility at each potential receptor.
6. Calculating annual dose (with a 95% confidence interval) from routine emissions to potentially the most exposed member of the public using the same model with the same assumptions each year so that potential doses from all years can be compared.
7. Calculating potential dose (with a 95% confidence interval) for each accidental release that was large relative to the routine emissions of the release year using the most current understanding of the behavior of tritium in the environment after an acute release.

This TDR for the Livermore site is comprised of five reports; this report summarizes those five.

- Part 1 describes DCART (**D**oses from **C**hronic **A**tmospheric **R**eleases of **T**ritium), the stochastic model (Peterson 2006) that was used to calculate dose from routine releases for the TDR.
- Part 2 (Peterson 2007a) presents data and supporting references primarily about past routine releases. It also provides a summary of historical tritium operations at the Livermore site. It lists the estimated annual release rates and uncertainties and explains the assumptions behind the derivation of the values. Other parameters (dilution factors, absolute humidity, relative humidity and mean annual observed concentrations of tritium in air at one location) and the assumptions behind derivation of their values and uncertainties are described.

¹⁵ The term “dilution factor” refers to the air concentration for unit source strength (or χ/Q); units are actually $\text{Bq m}^{-3}/\text{Bq s}^{-1}$ or $\text{Ci m}^{-3}/\text{Ci s}^{-1}$. The term, although standard for χ/Q , can be misleading because the higher the dilution factor, the higher the air concentration.

Part 3 (Peterson 2007b) presents two sets of doses with 95% confidence intervals predicted for routine releases from 1973 through 2005. These doses include the contribution from tritium released at SNL/CA from 1979 through 1995. One set was calculated from observed air concentrations at the VIS ambient air tritium monitor (adjacent to the LLNL Discovery Center – see Figure 1); the other set was calculated from release rates and dilution factors at the location of the SW-MEI (which was at the Discovery Center for most of the years). Both sets of doses were calculated using DCART. Doses to the SW-MEI predicted using DCART were compared to doses reported by LLNL in the SAERs, 1973 - 2005.

- Part 4 (Peterson 2007c) summarizes doses with 95% confidence intervals calculated using DCART at the locations of the SW-MEI from routine releases for 1953 – 1972 based on estimated annual release rates and dilution factors.
- Part 5 (Peterson 2007d) summarizes doses predicted to the MEI from accidental releases from the Livermore site in 1954, 1964, 1965, 1966, 1970, 1984, and 1985 and from SNL/CA in 1986 and 1987. These doses were calculated using an approach, described in the report, derived from the UFOTRI model.

This report, which is Part 6 of the TDR, summarizes the information contained in the first five extremely detailed reports. These reports describe all the assumptions that were made to carry out the TDR in its entirety. If new information were to be found that would add to what is contained in the reports or would contradict some of the assumptions of this TDR, it would be possible to revise the dose predictions accordingly.

It may be possible to confirm the doses estimated in this TDR by analyzing annual tree rings for OBT. If a suitable tree were found, the concentrations were above the detection limit of the analytical method, and enough wood were obtained for a measurable sample, the air concentrations to which the tree was exposed could be estimated. If this were to happen, doses could then be calculated from the estimated air concentrations using DCART.

Operations at LLNL's Livermore site began releasing tritium to the atmosphere shortly after the Laboratory was dedicated in September 1952. An estimated 792,000 Ci (29,300 TBq) of tritium had been released by the end of 2005 (the 95% confidence interval on this number is between 672,000 Ci [24,900 TBq] and 914,000 Ci [33,900 TBq]). 90% of this amount was released before 1973, and, of that, about 85% was released during two large accidental releases of 1965 and 1970 (Table 1).

For the years 1979 – 1995, when the Tritium Research Laboratory (TRL) at SNL/CA was routinely releasing environmentally significant amounts of tritium, tritium emissions from SNL/CA were included as a source of tritium for the TDR. Dose for the years 1973 – 2005, when the tritium concentration in air was monitored, was calculated from the observed air concentrations (which included the contribution from SNL/CA) as well as predicted from facility (i.e., LLNL and SNL/CA) release rates. An estimated 7,420 Ci (275 TBq) were released from the TRL, with a 2.5% confidence limit of 6,610 Ci (246 TBq) and a 97.5% confidence limit of 8,240 Ci (305 TBq); about 61% of the total tritium released was HTO. This total includes accidental releases of 200 Ci (7.4 TBq) of HTO in 1986 and 1,100 Ci (40.7 TBq) of HT in 1987.

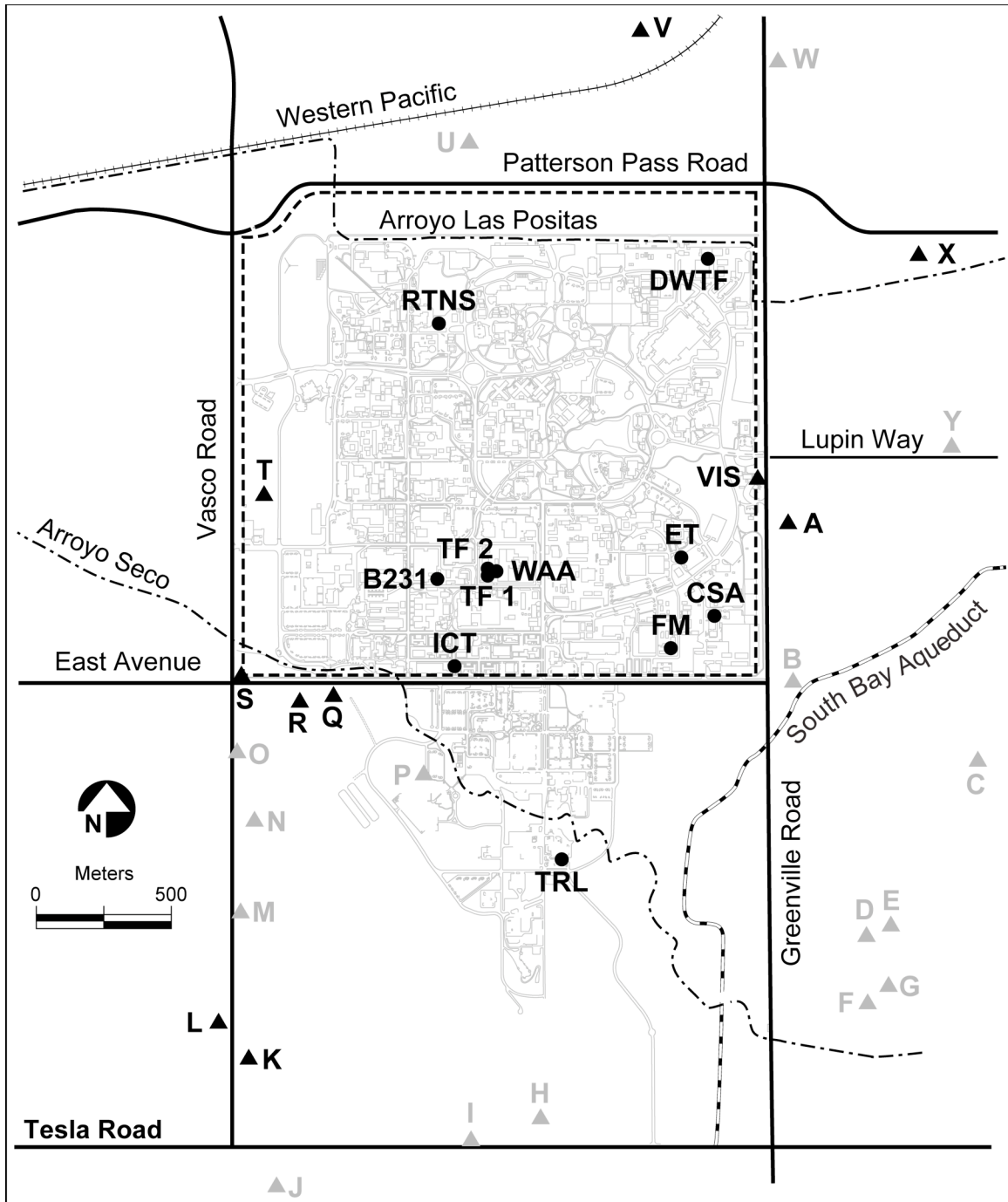


Figure 1. Sources of tritium (●) released from LLNL and SNL/CA relative to the locations (▲) of the site-wide maximally exposed individual (SW-MEI) (for routine releases) and the maximally exposed individual (MEI) (for accidental releases). Tritium was released routinely and accidentally from B231 (the early Tritium Facility), TF1 and TF2 (the south and north stacks of the current Tritium Facility), and TRL (Tritium Research Laboratory, SNL/CA). Routine releases were modeled from WAA (diffuse source at B331), ICT and room air (Building 212), RTNS (Building 292), CSA (Building 612 Yard and Building 624 incinerator), ET (evaporation trays), FM (Building 514 Yard), and DWTF (Decontamination and Waste Treatment Facility). A diffuse source was also modeled at B231. The SW-MEI was at Locations T (1953 – 1958), Q (1961 and 1974 – 1978), and VIS (all other years). The MEI was at Locations R (1954), Q (1964, 1966), X (1965), V (1970), A (1984), S (1985), L (1986), and K (1987). All marked locations were considered potential receptors.

Table 1. Curies of HT and HTO released from LLNL facilities on the Livermore site between 1953 and 2005. CL is the confidence limit on 95% confidence interval.

Type of release	Best estimate	2.5% CL	97.5% CL
Routine HT	104,000	90,300	119,000
Routine HTO	74,700	65,200	85,500
Accidental HT ^a	612,000	490,000	730,000
Accidental HTO ^b	1,450	597	2,300
Sources of routine releases			
HT from Tritium Facilities	95,900	82,500	110,000
HTO from Tritium Facilities	65,800	56,500	76,500
HT from other stacks	8,130	5,570	10,900
HTO from other stacks	2,410	1,900	2,940
HTO from area sources	6,470	5,500	7,490

Note: Because results from each category above involve sampling from different distributions, the 2.5% and 97.5% limits of the output distributions will be different.

^a Accidental releases of HT were from the current Tritium Facility, Building 331.

^b Accidental release of HTO was from Building 231 (which was called Building 102 at the time).

MATERIALS AND METHODS

Routine releases and annual dose calculations

Equilibrium dose model

The dose model used for the calculations was DCART (Peterson 2006). DCART was developed at LLNL to calculate doses for the TDR. DCART is a deterministic spreadsheet code in Microsoft Excel[®]. When coupled to Crystal Ball 2000^{®16}, a risk analysis software package that provides uncertainty and sensitivity analyses for spreadsheet codes, DCART produces probabilistic results. DCART is an equilibrium model that calculates annual doses (inhalation and ingestion of food and water) to adult, child (age 10 years), and infant (age 6 months to 1 year) from routine releases of HT and HTO to the atmosphere. Controlling input includes the HT or HTO annual release rates from each facility (in Ci y⁻¹) and dilution factors (χ/Q in s m⁻³) calculated by a dispersion model. Alternatively, mean annual measured concentrations of tritium in air can be used as the driving input. The concentration in soil water, due to the difficulty of accounting for emission of HTO from soil, is determined by empirical ratios between air moisture and soil water concentrations. For an HT release, HTO concentrations in air due to emission of HTO converted from deposited HT are estimated based on empirical ratios. The model calculates concentrations of tissue free-water tritium (TFWT or plant HTO) and OBT in edible plants (leafy vegetables, root vegetables, fruit or fruit vegetables [e.g., cucumber, tomato, eggplant, beans], grain, pasture, and hay) and animal products (milk, beef, pork, poultry and eggs). A balanced diet that provides the calories needed by an average adult, child, or infant is assumed.

Deterministic doses are calculated using dose coefficients for HTO and OBT (ICRP 1996) and HT (ICRP 1995) recommended by the International Commission on

¹⁶ Decisioneering, Inc. 1515 Arapahoe Street, Suite 1311, Denver Colorado 80202 USA.

Radiological Protection (ICRP); probabilistic doses are calculated from distributions determined by Harrison et al. (2002). The Harrison et al. mean values are higher than the ICRP values or those used in Federal Guidance Report 13 (Eckermann et al. 1999). Recommended parameter values and distributions of values for uncertainty analysis are specific to LLNL, to the extent possible. All input parameter values are distributed. Output for air concentrations and dose is reported as the mean and its 95% confidence interval. The model and its parameter values are described in detail in Part 1 of the TDR.

Predictions made using DCART of concentrations in air, vegetation, and animal products have been compared favorably with observations in various test scenarios of the International Atomic Energy Agency's (IAEA) coordinated research programs BIOMASS (BIOSpheric Modeling and ASSEssment) (IAEA 2003) and EMRAS (Environmental Modelling for RAdiation Safety¹⁷). Results of the tests are described in Part 1 of the TDR.

Model input

The parameters that can vary from year to year and are used to calculate annual dose in DCART include the release rate for each facility for each year, the dilution factor at the location of the SW-MEI for each facility¹⁸, the annual absolute humidity, and the annual relative humidity. Each of these values had an uncertainty applied to it. Distributions were usually normal for release rates¹⁹, normal for absolute and relative humidity, and lognormal for dilution factors.

Annual release rates

Data on release rates from facilities were collected. Numerous facilities have released small quantities of tritium throughout the years, but if the major contributors are accounted for and if the uncertainties applied to the release rates are generous enough, all contributions of tritium to dose will be taken into account. The major facilities with stack releases that were modeled for the TDR and the best estimates of their routine releases are listed in Table 2²⁰.

Confidence is high on the reported release rates from stacks after 1973, when the release rates from Building 331 were speciated into HT and HTO. From mid-1956 through 1972, confidence on the total tritium released from the site is reasonably high because releases were documented in the quarterly reports to the AEC. From the inception of the Laboratory until mid-1956, assumptions about release rates with high uncertainty were made based on a memo by J.L. Olsen of LLNL to the AEC (Olsen 1973) that summarized releases from the Tritium Facility (which was Building 231 from 1953 through 1958 and Building 331 thereafter) and Building 212 for the years that the lab was operational. In the Olsen memo, annual release rates from 1953 through 1963 were reported with $\pm 50\%$ uncertainty. After comparing the release rates in the quarterly memos between 1956 and 1963 to the values in the Olsen memo, the values in the Olsen memo were adjusted to

¹⁷ A report will be issued after EMRAS terminates in the fall of 2007. Information may be found at <http://www-ns.iaea.org/projects/emras/>.

¹⁸ Or location VIS, when predictions were compared with observations.

¹⁹ The value of the release rate was always left-truncated at zero if the uncertainty was large.

²⁰ Emissions from the incinerator at Building 624 (1977 – 1988) and emissions from the Decontamination and Waste Treatment Facility (2004 and 2005) were also modeled because stack parameters were known, but their impact on dose was very small.

scale according to the difference between the Olsen memo and the quarterly reported releases. Additional uncertainty was applied to the release rates to account for this adjustment.

Table 2. Major facilities with stack releases, years of operation, and estimated quantities (Ci) of HT and HTO released routinely. Buildings are shown with the current numbering system. Identifiers for Figure 1 are shown in parentheses.

Building	Years of operation	HT	HTO
212 ^a – accelerators (ICT)	1953 - 1987	7,470	1,050
231 – early Tritium Facility (B231)	1953 - 1958	35,000 ^b	3,500 ^b
292 – Rotating Target Neutron Source (RTNS)	1979 - 1989	-----	1,220
331 – Tritium Facility (TF)	1959 - present	38,300	59,200
TRL of SNL/CA (TRL)	1979 – 1995	1,590	4,540

^a Between 1953 and 1967, tritium was released from the Cockcroft-Walton Accelerator and from the 90-inch cyclotron to room air (modeled as a stack release); in 1967, the Insulating Core Transformer Accelerator began operations and tritium releases were through a stack.

^b Because the speciation of these releases was unknown, doses were also calculated using the conservative assumption that 18,000 Ci of HT and 21,000 Ci of HTO were released (i.e., 54% of the total tritium released was HTO).

It was important to accurately assess (with appropriate uncertainty) the fraction of the total release that was either HT or HTO because, as modeled in DCART, a 1 Ci release of HT will have only about 5% the dose impact of a 1 Ci release of HTO, all other factors being the same. After 1973, when releases from the Tritium Facility were measured as HT and HTO, the uncertainty was minimal. However, prior to that, assumptions about the fractions of HT or HTO had to be made.

According to the records (See Table 1 in Part 2 of the TDR), tritium releases in the early years were fairly large puffs of HT (> 100 Ci) on a very few specified dates in any one year. Most probably, the large puffs that were released in the years before 1961 were 100% HT, but to be conservative, one set of doses was calculated based on the assumption that the puffs were about 10% HTO. To be even more conservative²¹, an additional set of doses was calculated assuming that the puffs were 54% HTO. The assumption that the releases might be as much as 54% HTO was made based on molecular sieve sampling data from the Tritium Facility after 1973. These sampling data demonstrated that when the total activity of tritium released annually from one stack was greater than 630 Ci, the percentage of the total that was HTO averaged 54%. They also demonstrated that when less total tritium is released annually per stack, the fraction of the total release that is HTO rises²².

From 1961 through 1972, when release rates were only reported quarterly and speciation of releases at the Tritium Facility was not yet determined, the only assumption applied was that 54% of the release was HTO. Thus the upper confidence limits (UCL) on the second set of dose predictions for 1953 through 1960 are directly comparable to the UCL on predictions from 1961 through 1972.

²¹ Probably overly conservative.

²² Release rates per stack exceeded 630 Ci per year prior to 1973 with the exception of Stack 1 in 1972, when 506 Ci were released and 64% of the release was assumed to be HTO.

The uncertainty applied to the release rates and speciation changed from year to year. Assigned uncertainty on both was as high as $\pm 40\%$ in the early years.

Uncertainty that was common to stacks, stack sampling, and ambient air sampling includes

- Accuracy of ion chambers ($\pm 14\%$)
- Similarity between sampled stack air and exhausted stack air ($\pm 10\%$)
- Calibrated exit velocity of the stack ($\pm 5\%$)
- Flow rate through stack and ambient samplers estimated from rotameter readings ($\pm 14\%$)
- Silica gel correction factor prior to 2001 ($1.6 \pm 15\%$) (Guthrie et al. 2002; Peterson 2007a)

Other uncertainties varied depending upon the facility. These included analytical uncertainty on the tritium samples (usually very small) and the uncertainty about the exit velocity for the years that flow rate was not measured. For Building 331, uncertainty had to be added ($\pm 28\%$) when, between 1962 and 1972, the records did not document from which of the two stacks the tritium was released.

These uncertainties (release rates, speciation, and stack sampling) resulted in uncertainty estimates on release rates that ranged from $\pm 18\%$ to $\pm 61\%$ for Building 331 and even as high as $\pm 71\%$ for Building 212 in the early years.

Tritium has also been released to the atmosphere from diffuse area sources. Diffuse sources included areas where tritiated waste or contaminated equipment was stored prior to disposal. Area sources are important to dose close to the Laboratory because the tritium that diffuses from them is not diluted by large volumes of stack flow. At nearby locations, the dose impact of a relatively small quantity of tritium from a diffuse source can be much higher than the same quantity released from a stack. Since 1992, the sources of diffuse tritium were documented and annual release rates from them estimated. For the TDR, because of the large impact on dose, assumptions had to be made to account for the potentially important dose contribution from sources whose existence or release rate was unknown. The assumed and actual area sources are listed in Table 3.

The uncertainty applied to the release rates from diffuse sources approached and exceeded $\pm 100\%$ in those cases when the existence, location, and release rate of the source were being assumed. Even this amount of uncertainty probably does not address the true uncertainty, but assumptions about the magnitude of the release rates were very conservative to account for all potential diffuse sources and to assure that doses would not be underestimated. Thus the relatively small uncertainty of $\pm 100\%$ about the release rate should nevertheless include the maximum likely release rate, although the lowest likely release rate could easily have been grossly overestimated.

A comprehensive description of documented release rates and a discussion of the derivation of release rates and associated uncertainties may be found in Part 2 of the TDR.

Table 3. Locations of assumed and actual area sources, years of operation, and estimated quantities (Ci) of HTO released routinely. Buildings are shown with the current numbering system. Identifiers for Figure 1 are shown in parentheses.

Building	Years of operation	HTO
Building 231 (B231)	1953 - 1958	800
Building 331 (WAA)	1959 - present	3,000
Building 514 (FM)	1953 - 1961	200
Building 612 (CSA)	1965 - present	2,000
Evaporation Trays (ET)	1962 - 1976	500

Routine dilution factors

In DCART, concentrations in air are calculated by multiplying a release rate by a dilution factor. Dilution factors were calculated using the regulatory dispersion and dose model, CAP88-PC (Parks 1992), and a generic wind file representing 15-minute data obtained from the LLNL meteorological tower for 2000 – 2003. This wind file was used for all calculations, because, once uncertainty associated with dispersion is accounted for, there is very little difference year-to-year in the LLNL wind profile. Comparison of air concentrations and doses is therefore straightforward because the only variables affecting them will be the release rates from the facilities and any changes in stack parameters. Dilution factors, of course, are specific from a source to a receptor and will change if the receptor changes. Table 4 shows representative dilution factors²³ from various facilities to the location of the Discovery Center at LLNL. Although strongly influenced by the CAP88-PC wind file, the dilution factor can be seen to depend on distance and direction from the source to the receptor and to whether the effective release height was high or low. A complete list of dilution factors at the Discovery Center is found in Part 2 of the TDR.

For each year of the TDR, the location of the SW-MEI had to be assessed based on the likelihood that a person could have lived at a particular location and the magnitude of the air concentrations predicted from release rates and dilution factors derived for each facility for that location. The SW-MEI was determined to have been at Location T from 1953 – 1958, at Location VIS for the years 1959, 1960, 1962 – 1973, and 1979 – 2005, and at Location Q in 1961 and 1974 – 1978 (Figure 1). The dilution factors for Location Q are found in Parts 3 and 4 of the TDR; the dilutions factors for Location T are found in Part 4 of the TDR.

The stack parameters (height, inside diameter, and exit velocity), and the directions and distances from source to receptor, needed as input to CAP88-PC to derive the dilution factors at all potential locations of the SW-MEI are found in Parts 2, 3, and 4 of the TDR.

An uncertainty of ± 25 or 30% on the dilution factor is mainly due to the expected uncertainty on a Gaussian dispersion model with the proper input parameters, which is a factor of two to four over flat terrain, with accuracy decreasing as complexity of meteorological and terrain conditions increases (Miller and Hively 1987). Additional uncertainty on the dilution factor is added by accounting for the uncertainty on the exit

²³ When stack parameters, particularly flow rate, change, the dilution factor will change.

velocity. In the case of Building 231, when nothing except the diameter of the stack could be documented, the uncertainty applied to the exit velocity was about $\pm 30\%$ at Location T, and the overall uncertainty on the dilution factor was $\pm 43\%$ ²⁴.

Table 4. Representative dilution factors used in DCART to predict air concentrations and doses at the Discovery Center. Uncertainty on these numbers ranges from 25 – 40%.

Facility	Dilution factor (λ/Q); s m ⁻³
Stack sources	
Building 212	2.494 10 ⁻⁶
Building 231	2.389 10 ⁻⁶
Building 292	4.425 10 ⁻⁷
Building 331 south	1.220 10 ⁻⁶
Building 331 north	1.084 10 ⁻⁶
SNL/CA TRL stack	6.595 10 ⁻⁷
Diffuse sources	
Building 231	2.758 10 ⁻⁶
Building 331	3.474 10 ⁻⁶
Building 514	1.076 10 ⁻⁵
Building 612	1.763 10 ⁻⁵
Evaporation trays	2.008 10 ⁻⁵

The method by which dilution factors were derived is described in Part 2 of the TDR.

Absolute and relative humidity

Absolute humidity is used in DCART to calculate air moisture concentrations (Bq L⁻¹) from concentrations of tritium in air (Bq m⁻³)²⁵. It is necessary to calculate air moisture concentrations before calculating concentrations in plants and animals (see Part 1 of the TDR). Relative humidity is needed to calculate the TFWT concentrations in plants.

Annual absolute humidity for 1973 through 2005 was calculated from the ratios of the grams of water collected biweekly by the silica gel ambient air tritium samplers divided by the air flow calculated or measured through the samplers for the same sampling period. When mean annual absolute humidity calculated in this manner was compared with mean annual absolute humidity values obtained from data collected at the LLNL meteorological tower between 1999 and 2003, the silica gel estimates were on average 4% lower, but a consistent underestimation of absolute humidity by the silica gel method was not shown. Underestimating absolute humidity in DCART results in a small increase in dose. For the dose calculations from 1953 through 1972, the mean of all observed absolute humidity values was used; uncertainty on all values was $\pm 5\%$. The uncertainty applied to the mean annual relative humidity (obtained from LLNL meteorological data) was $\pm 10\%$. Values for absolute humidity and relative humidity used in the TDR are listed in Part 2 of the TDR.

²⁴ This percent uncertainty is slightly higher than the highest uncertainty at the Discovery Center (Table 4).

²⁵ Bq L⁻¹ = Bq m⁻³ / L H₂O m⁻³

Observed concentrations of HTO in air

Measured mean annual concentrations of HTO in air (Bq m^{-3}) can be used directly as input into DCART to calculate doses that should be inherently more accurate (although not necessarily less uncertain) than even the best of predictions based on release rates and dilution factors. The uncertainty associated with the observed concentrations of HTO in air volume included the collection efficiency of the silica gel, the uncertainty on the analytical results, the uncertainty due to missing samples or samples below the detection limit, and the uncertainty on the volume of air that passed through the samplers.

VIS was chosen as the air tritium sampling location at which to calculate tritium concentrations in air and air moisture (and doses) for this TDR because of its proximity to the UNCLE Credit Union, the location of the Livermore site's SW-MEI for NESHAPS²⁶ compliance and because air tritium sampling has been carried out at VIS since 1973. If predicted air concentrations could be shown to equal or exceed the observed air concentrations at this location (which turned out to be the SW-MEI for 1959, 1960, 1962 – 1973, and 1979 – 2005), more confidence could be placed in the predictions for the years without any ambient air sampling (i.e., 1953 – 1972).

Model calculations and comparisons

Doses to the SW-MEI were calculated using source terms and dilution factors in DCART for all years from 1953 through 2005. The SW-MEI for each year was selected using dispersion modeling to determine the air concentrations at potential locations of the SW-MEI, which were selected from United States Geological Survey (USGS) topographic maps on which structures were indicated. Any structure near the Livermore site was assumed to be a residence. When the Discovery Center area was the potential location of the SW-MEI, it was assumed to be a residence even in those years when the Discovery Center itself did not exist.

Doses and their 95% confidence intervals were also calculated using the observed mean annual HTO concentrations in air at VIS for 1973 through 2005. The resulting doses were compared with doses predicted from release rates and dilution factors. A similar comparison was made between air moisture concentrations predicted by DCART using release rates and dilution factors and observed mean annual air moisture concentrations at VIS. This comparison was essentially a test of the dispersion model and the assumptions made for the TDR. Uncertainties in both observations and predictions were taken into account.

Probabilistic doses predicted using DCART with release rates and dilution factors as input were also compared with the deterministic doses reported by LLNL in the SAERs from 1973 through 2005.

²⁶ National Emissions Standards for Hazardous Waste Pollutants, 40 CFR 61 Subpart H. (National Emission Standards for emissions of Radionuclides other than Radon from Department of Energy Facilities).

Accidental releases and dose calculations

Time-dependent model

Modeling dose after an acute release of tritium requires a tritium-specific time-dependent model that accounts not only for dispersion of the plume but also for the changing relationship between wind speed and stability class and the uptake of HTO by plants and production of OBT. For example, conditions that will result in a low inhalation dose may be the best for uptake of tritium by plants and, as a consequence, a high ingestion dose. The model also must account for the deposition of HT or HTO and the (re)emission of HTO from the soil and its subsequent dispersal and uptake by plants. Ingestion dose is potentially the most important dose after an accidental release of tritium, particularly a release of HT.

There are only a handful of models in the world that model dose after an acute tritium release. The most successful, most established, most tested, and best known of these models is UFOTRI (Raskob 1990, 1993). Even with UFOTRI, however, it is impossible to model accidental releases without having at least hourly meteorological data for several days after the release. Hourly data allow the model to account for shifts in wind direction, wind speed and stability, all of which impact the air concentrations at particular locations and the uptake by plants. Detailed meteorological data were unavailable except for the most recent (and inconsequential) accidental releases, so an alternative approach to modeling them was needed.

An equilibrium model, such as DCART, may be able to calculate a dose equal to that calculated using a time-dependent model because the dose integral over infinite time per unit of release is numerically equal to the dose at a future steady-state when that release is repeated indefinitely at unit rate (Barry 1979). In other words, if it is assumed that the accidental release is spread out over the year and that the winds during that year blow only in one direction (the dilution factor being calculated using a dispersion model for acute releases), then the integrated air concentration for a year to which the individual is exposed is the same as that to which the individual was exposed during the accidental release.

By testing DCART in a hypothetical HT or HTO accident scenario in EMRAS, it was found that DCART could predict the inhalation dose after an accidental release of HTO as accurately as any of the participating models, but it overestimated the inhalation dose after an accidental release of HT. Furthermore, the ingestion dose predicted by DCART was low, particularly for the HT release. Apparently, the potential equivalency between equilibrium and dynamic predictions of ingestion dose does not hold for tritium.

When modeled dynamically, predicted inhalation dose and predicted ingestion dose are correlated, although the relationship will vary depending upon the meteorological conditions (e.g., wind speed, stability class and whether it was raining) and whether the release was HT or HTO. It thus seemed reasonable to use an HTO inhalation dose predicted by DCART and what is known about ingestion/inhalation ratios after an acute release of HTO to calculate potential ingestion dose for an HTO release. Similarly, what is known about the ratio of acute-release-HT inhalation dose to acute-release-HTO inhalation dose, given the same release rates and meteorological conditions, can be used to convert HTO inhalation doses predicted by DCART to HT inhalation doses that can

then be used with what is known about ingestion/inhalation dose ratios after an acute release of HT to calculate ingestion dose for an HT release.

UFOTRI was used to calculate potential doses at the three locations competing for ITER, the international experimental fusion reactor (Raskob et al. 1999). These calculated inhalation and ingestion doses from acute releases of HT and HTO were a starting point in the development of the ingestion/inhalation ratios for this TDR. With considerable guidance from UFOTRI's developer, Dr. Wolfgang Raskob of Forschungszentrum Karlsruhe GmbH, Karlsruhe, Germany, that drew heavily on his experience and expert knowledge, a method (see Part 5 of the TDR) was developed whereby doses for all accidental releases could be calculated from a distributed HTO inhalation dose calculated by DCART.

Source terms for accidental releases

Because of the extreme difficulty of modeling dose after an acute release of tritium (see Appendix B of Part 5 of the TDR), all accidental releases that were a relatively small fraction of the annual total tritium released the year of the incident were included in the estimated annual routine releases that were modeled for the TDR. However, there were seven accidental releases from LLNL and two from SNL/CA that were relatively too large to be modeled as routine. The LLNL accidental releases, with their uncertainty and type of distribution, included

- October 13, 1954: $1,450 \pm 435$ Ci (53.7 ± 16.1 TBq) HTO; lognormal (Stanhope, 1954)
- 1964²⁷ (date unspecified): $0 - 24,000$ Ci ($0 - 888$ TBq) HT; uniform (Souers 1988; 2006)
- January 20, 1965: $207,000 - 309,000 - 360,000$ Ci ($7,660 - 11,400 - 13,300$ TBq) HT; triangular (Peterson et al., 2002)
- April 7, 1966: $12,500 \pm 3,090$ Ci HT (463 ± 114 TBq); lognormal (May 1966)
- August 6, 1970: $288,000 \pm 51,600$ Ci ($10,700 \pm 1,910$ TBq) HT; lognormal (Myers et al., 1973)
- June 8, 1984: $5,510 \pm 1,110$ Ci (204 ± 41.1 TBq) HT; lognormal (Hill 1984; Howe 1985a)
- January 24, 1985: 972 ± 182 Ci (36 ± 6.73 TBq) HT; lognormal (Howe 1985b)

The 1954 release was from what is now Building 231, which is where tritium operations were carried out before the present Tritium Facility (Building 331) was built in 1959. All other LLNL accidental releases were from Building 331.

SNL/CA had two accidental releases from the TRL that could not be modeled as routine. One was a release of 200 ± 20 Ci HTO that probably occurred on January 11, 1986 (Garcia and Gorman 1996; Hafner 2006); the second was a release of $1,100 \pm 55$ Ci HT that occurred on August 18, 1987 (Garcia and Gorman 1996; Garcia 2006).

²⁷ It is likely this release did not happen, but it has been included to be health-protective. See discussion in Part 5 of the TDR.

Dilution factors for accidental releases

HOTSPOT (Homann 1994) was the dispersion model of choice to estimate dilution factors for the accidental releases, because it is an accident consequence model that has been tested and verified and needs minimal input data. Different methods were used to estimate the dilution factors depending upon how much information was known about the releases. When release times were unknown (as in 1954, 1964, and 1966), a set of wind speeds occurring with 10%, 50%, and 90% probability were obtained for each stability class from 15-minute data from the LLNL meteorological tower for six years for the month of the release²⁸ (hourly data were used for the entire year in 1964 because even the month of this release was unknown²⁷). These combinations of wind speed and stability class were used as input to HOTSPOT to obtain a distribution of dilution factors. For the large accidental releases of 1965 and 1970, observed wind speed and stability class had been recorded and was used as input to HOTSPOT. For the releases in 1985, 1986, and 1987, the archived LLNL 15-minute data were used in HOTSPOT for the duration of the releases. The meteorological data for the time of the June 1984 release were missing, so a generic wind file was developed.

The minimal uncertainty on the dilution factors for the accidental releases was $\pm 80\%$ based on the uncertainty needed to contain the air concentrations predicted by eight modelers participating in the EMRAS hypothetical scenario²⁹ and to be conservative. The uncertainty for the 1954 and 1970 releases was as high as $\pm 200\%$. Potential locations of the MEI were selected from USGS topographic maps on which structures were indicated. Any structure was conservatively assumed a potential residence, and a dilution factor was calculated for the closest structure downwind of the accident (or simply the closest structure to the release point when wind direction was unknown). Each potential structure was provided with x- and y-coordinates in HOTSPOT on a grid with the source at the center³⁰.

Dilution factors varied by about a factor of 60. They were lowest (about $1 \times 10^{-6} \text{ s m}^{-3}$) for the LLNL release of 1965 and the SNL/CA release of 1986, when meteorological data were well known, and highest in 1954 (about $6.3 \times 10^{-5} \text{ s m}^{-3}$), when meteorological data were unknown, the effective release height was low, and the location of the MEI was chosen to be the closest structure to the source. Dilution factors for 1964, 1966, and 1985 were similar (about $6.5 \times 10^{-6} \text{ s m}^{-3}$), probably due to the method used to estimate the dilution factor without knowing the time of the release and the assumed locations of the MEIs. Dilution factors were also similar for 1970, 1984, and 1987 (about $2 \times 10^{-6} \text{ s m}^{-3}$) when meteorological data were well known.

Dilution factors were estimated for all the potential locations of the MEI shown in Figure 1 to determine those locations having the highest values (i.e., doses). The MEIs

²⁸ Wind conditions are seasonal, and compiling data for a month for six years provides a reasonable generic profile. An extended workday was assumed conservatively to include the light winds and stable conditions associated with early morning and evening conditions that would result in higher doses.

²⁹ The HOTSPOT manual states, "Given accurate input assumptions, the standard deviation of the dose values [i.e., dilution factors] as calculated in Hotspot is approximately a factor of 5. Therefore, 68% of the time (i.e., the percentage of observations within 1 standard deviation, assuming a Gaussian distribution), the calculated dose values will be within a factor of 5."

³⁰ To assume that the plume blew directly at the structure is unrealistic.

were assumed to be at Locations R (1954), Q (1964, 1966), X (1965), V (1970), A (1984), S (1985), L (1986), and K (1987).

Dose calculations for accidental releases

Distributed dilution factors were used along with distributed release rates in DCART to obtain distributed HTO inhalation doses (assuming all releases were HTO). Ingestion dose was calculated in Microsoft Excel[®] coupled to Crystal Ball[®] using an equation described in Part 5 of the TDR that converts the HTO inhalation dose predicted by DCART to an inhalation and ingestion dose for an acute release of HT and an ingestion dose for an acute release of HTO. Total dose is calculated by summing the inhalation and ingestion doses.

All parameter values in the equation are distributed. In addition to the inhalation/ingestion ratios derived from experience with UFOTRI, other parameters are used in the equation to reduce the ingestion dose to better apply to the Livermore environment and yet remain conservative. In UFOTRI, as much as 82% of the ingestion dose is attributable to grain. This potential contribution was removed from the dose calculations for the accidental releases, because, although the Livermore Valley was once noted for its grain production, grain was not being grown at the locations of the potential MEIs; the doses predicted using this assumption are conservative maxima. A further reduction in dose was made based on the probability that edible plants were growing at the time of the accidental release; the doses predicted using this assumption are realistic. Other than these changes, the ingestion dose pathway derived from UFOTRI is, as in DCART, based on an entire, complete diet assumed contaminated to the maximum extent possible at the location of the predicted air concentration. Detail about the derivation of the parameter values is provided in Part 5 of the TDR.

The total dose was predicted with 95% confidence intervals (i.e., 2.5% and 97.5% confidence limits) for all accidental releases. In addition, doses with 90% confidence intervals (i.e., 5% and 95% confidence limits) were calculated for the accidental release in 1970 so that the dose predictions of the TDR could be compared with those of the Agency for Toxic Substances and Disease Registry's (ATSDR) Public Health Assessment (PHA) (ATSDR 2003).

RESULTS

Predicted vs. observed concentrations of HTO in air moisture

Predicted and observed concentrations of HTO in air moisture (Bq L^{-1}) at VIS from 1973 through 2005 are compared in Figure 2.

Predicted air moisture concentrations were higher than those observed at VIS for all years except 1974. The UCLs for the predictions barely exceeded the UCLs for the observations for 1986 (Figure 2), however, due to especially high uncertainty about the 1986 observations. Only six years (1981, 1989, 1991, 1995, 1998, and 1999) were overestimated to the extent that the confidence intervals of predictions and observations showed no or very little overlap. These years without overlap correspond to predicted-to-observed (P/O) ratios for the means that were greater than a factor of 2.

The size of the confidence intervals, defined as the UCL divided by the lower confidence limit, is reasonably small for both observations and predictions. The mean interval on the predictions is a factor of 2.7, slightly greater than the factor of 2.3 for the observations; the maximum interval on the observations was a factor of 4.8 in 2005; the maximum on the predictions was a factor of 3.8 in 2000.

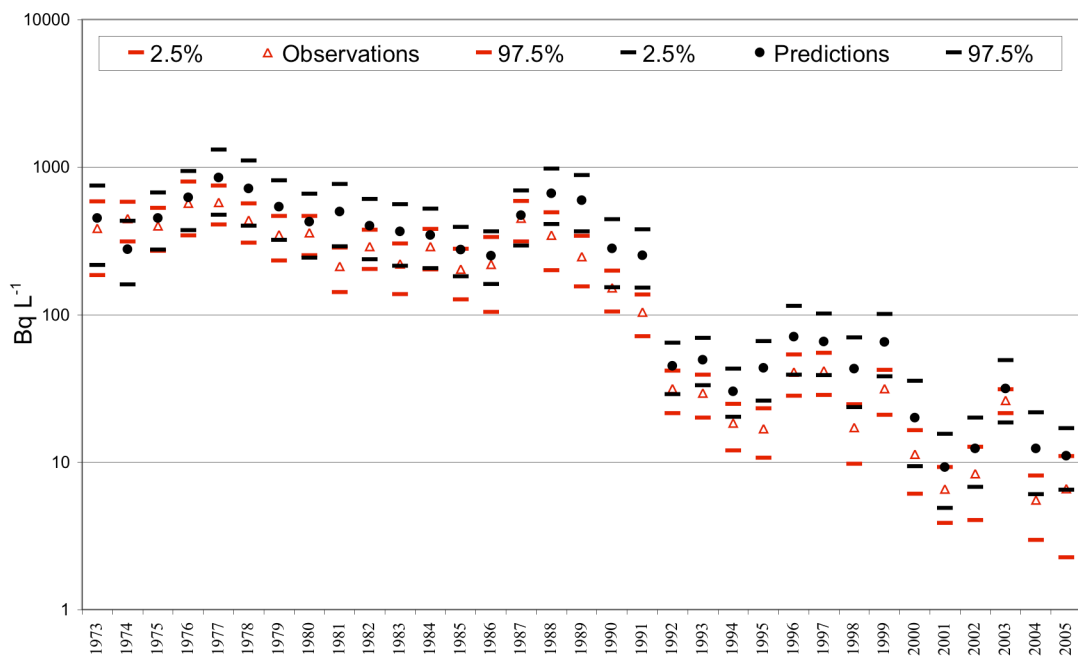


Figure 2. Mean predicted and observed tritium concentrations in air moisture at VIS with 95% confidence intervals on both. Predictions were calculated using CAP88-PC and observed mean annual absolute humidity (AH). Values for AH were calculated from the water retained on the silica gel and the air flow through the sampler.

Annual dose predictions

The complete dose history to the SW-MEI for routine releases from LLNL (that includes the contribution from SNL/CA between 1979 and 1995) is shown in Figure 3. All predictions in Figure 3 were calculated using DCART at the locations of the SW-MEI that had been determined using dispersion modeling. The predicted annual doses from routine releases shown in Figure 3 are not completely comparable for all years. There are three main reasons for this: one is that the location of the SW-MEI varied depending on the location of the dominant emission source, another is the effect of assuming 54% of the releases were HTO in the years 1953 through 1960, and the third is the fact that doses were calculated when possible using observed, rather than predicted, concentrations of tritium in air. The reasons for the lack of comparability are discussed in turn.

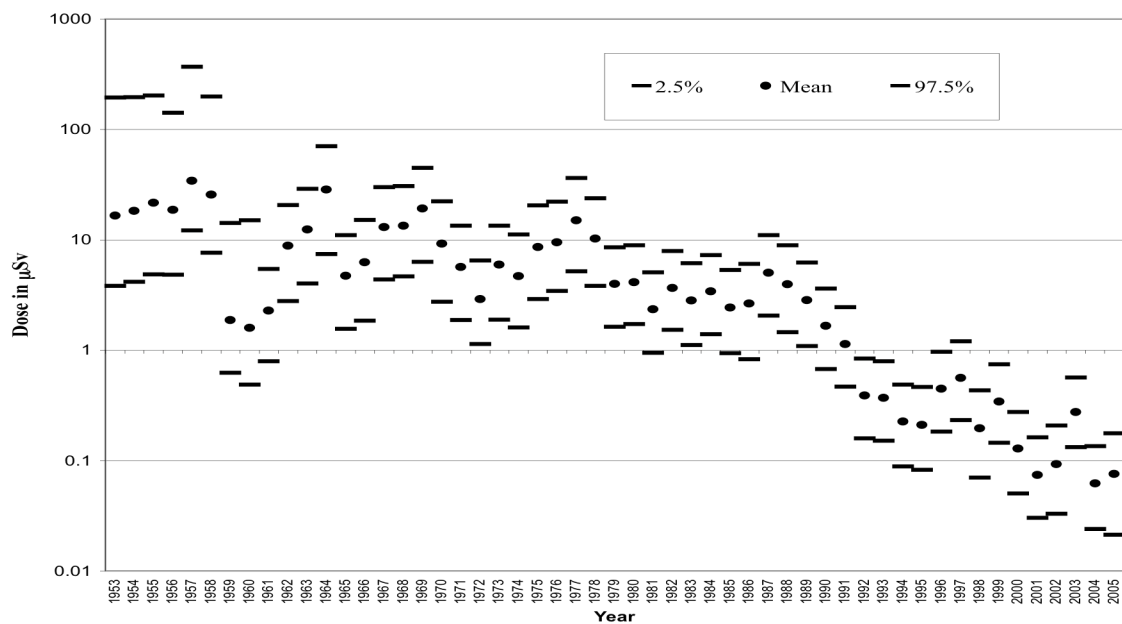


Figure 3. Annual doses (with 95% uncertainty intervals) predicted by DCART to the SW-MEI from routine LLNL (and SNL/CA from 1979 – 1995) tritium releases. The SW-MEI was at Location T from 1953 – 1958, at Location Q in 1961 and 1974 – 1978, and at Location VIS in 1959, 1960, 1962 – 1973, and 1979 – 2005. Doses were predicted from release rates and dilution factors except for 1973 and 1979 – 2005 when doses were predicted using the observed mean annual concentrations of HTO in air at Location VIS.

Potential locations of the SW-MEI included P, Q (after 1958), R, S, T, and VIS. The SW-MEI was determined to have been at Location T from 1953 through 1958, at Location Q in 1961 and from 1974 through 1978, and at Location VIS in 1959 and 1960³¹, from 1962 through 1973, and from 1979 through 2005. Doses were calculated using release rates and dilution factors for all years except those when the SW-MEI was determined to have been at the Discovery Center, the location of an ambient air tritium monitor (VIS). For these years, doses were calculated from mean annual observed HTO concentrations.

For 1953 through 1958, the uncertainty intervals on the doses for all potential locations of the SW-MEI overlapped, but the dose at Location T was about 50% higher than at Locations R and S and more than a factor of three higher than at Locations P and VIS. Dose at Location T was about 30% higher than at Location Q in 1958.

In 1959 and 1960, when VIS was determined to be the location of the MEI, there was only about 10% difference between the dose at VIS compared with that at Location Q. In 1961, Location Q edged out VIS by about 30% to become the location of the SW-MEI, although there was not more than a factor of two difference between the doses calculated for all the potential locations. The SW-MEI shifted back to the Discovery Center (VIS) from 1962 through 1973. For these years, dose at the Discovery Center was about 25 – 50% higher than at Location Q, but all doses for potential locations of the SW-MEI were

³¹ In 1959 and 1960, the predicted mean doses (assuming about 10% of the tritium released from Building 231 was HT) at Location T were the same or higher than those at Location VIS. Location VIS, however, was selected as the SW-MEI because the UCL limits on the doses at Location VIS were higher than those at Location T.

no more than a factor of two lower. The situation was similar when the SW-MEI was at Location Q between 1974 and 1978. Doses at Location Q were then about 20% higher than those at VIS. From 1979 onwards, the SW-MEI was at the Discovery Center (i.e., Location VIS). Because the land on which Locations Q, R, and S stood was purchased in 1986 or 1987 to add to SNL/CA's acreage, these locations could no longer be considered as potential SW-MEIs, i.e., they were no longer in potential residential use.

Because the speciation of releases into HT and HTO during the early years of Laboratory operations was not quantifiable (see Part 2 of the TDR), two different sets of assumptions were used to calculate doses for 1953 through 1960. The confidence limits shown in Figure 3 for 1953 through 1960 were derived from both sets of assumptions: the 2.5% confidence limit and mean were calculated using the assumption that the tritium released from either Building 231 or 331 was about 10% HTO (actually 5 to 13% depending upon the year), while the 97.5% confidence limit (i.e., the UCL) was calculated using the assumption that 54% of the total tritium released was HTO. The UCL based on the assumption of 54% HTO is a factor of between about 3 and 4.5 times higher than the UCL based on the assumption that about 10% of the release was HTO (not shown in Figure 3). Until 1960, the uncertainty on the predictions (UCL assuming 54% HTO divided by the lower confidence limit assuming 10% HTO) averaged a factor of 35. Between 1961 and 1972, the average uncertainty was a factor of 7.3, and after 1973 the mean uncertainty on the dose predictions was a factor of 5.6.

The highest mean doses from routine releases were in 1957 (34 μSv [3.4 mrem]), 1964 (29 μSv [2.9 mrem]), 1958 (26 μSv [2.6 mrem]), and 1955 (22 μSv [2.2 mrem]). Doses dropped by an order of magnitude between 1959 and 1961, but rebounded in 1962. They remained within about a factor of three on either side of 10 μSv (1 mrem) from then until 1992 when doses dropped significantly. The mean dose in 1960 was the lowest until 1991.

Doses compared in the figure are to an adult member of the public, which is the normal receptor for regulatory purposes. Even with the high uncertainty applied to the early years, the UCL on the predictions only exceeded 100 μSv (10 mrem) for the first six years, with the highest UCL being 370 μSv (37 mrem). Doses were about a factor of 10 higher, on average, from the early 1970s through 1991 compared with doses after 1992. The UCLs on all doses after 1958 were below the present 100 μSv (10 mrem) annual dose limit for NESHAPs compliance for dose from atmospheric releases.

Doses from 1973 through 2005 were calculated at the Discovery Center using the ambient air data obtained at VIS (all except the results for 1974 through 1978 are shown in Figure 3). A comparison of these doses and those predicted from releases from facilities results in a figure very much like Figure 2, in which predicted and observed HTO concentrations in air moisture were compared. The figure (not shown; see Figure 11 in Part 3 of the TDR) is not identical, however, because the observed input to DCART was in different units³² (in Bq m^{-3} rather than Bq L^{-1} , the units in Figure 2) and because there is considerable overlap between the confidence intervals of both sets of doses

³² The relationship between doses predicted from release rates or from ambient air concentrations should be similar to the relationship between predicted and observed air concentrations (Figure 2), and it is except for 1973 and 1976. For those years (see Part 2 of the TDR), the linear relationship that exists between tritium concentrations in air moisture (Bq L^{-1}) and in air volume (Bq m^{-3}) did not hold – the concentrations in air volume were higher than expected.

because the uncertainty on predicted doses is higher than that on either predicted or observed air concentrations.

The UCL for doses predicted from release rates was higher than that from doses predicted from observed air concentrations except for 1973, 1974, and 1976, but the overlap in the confidence intervals on the two sets of dose predictions is such that they may be considered the same. The highest dose predicted at the Discovery Center after 1973 from release rates had an UCL of 22.3 μSv (2.23 mrem) in 1977, while the highest dose predicted from ambient air concentrations had an UCL of 19.7 μSv (1.97 mrem) in 1976.

LLNL historical annual dose predictions compared with those of the TDR

In Figure 4, doses and the 95% confidence intervals predicted by DCART to the SW-MEI from release rates and dilution factors are compared with tritium doses reported by LLNL in SAERs between 1973 and 2005. “CAAC” is the Clean Air Act Code (Moore et al. 1979), or AIRDOS-EPA, that became CAP88-PC (Parks 1992), the code used for LLNL’s compliance with NESHAPs. “CPS” is the Continuous Point Source Code (Peterson et al. 1976). “Other” refers to the use of the highest observed air concentration to calculate dose in 1973 and the use of a dispersion equation in 1974 to calculate air concentration. Note that use of the CPS code and CAAC overlapped between 1986 and 1991.

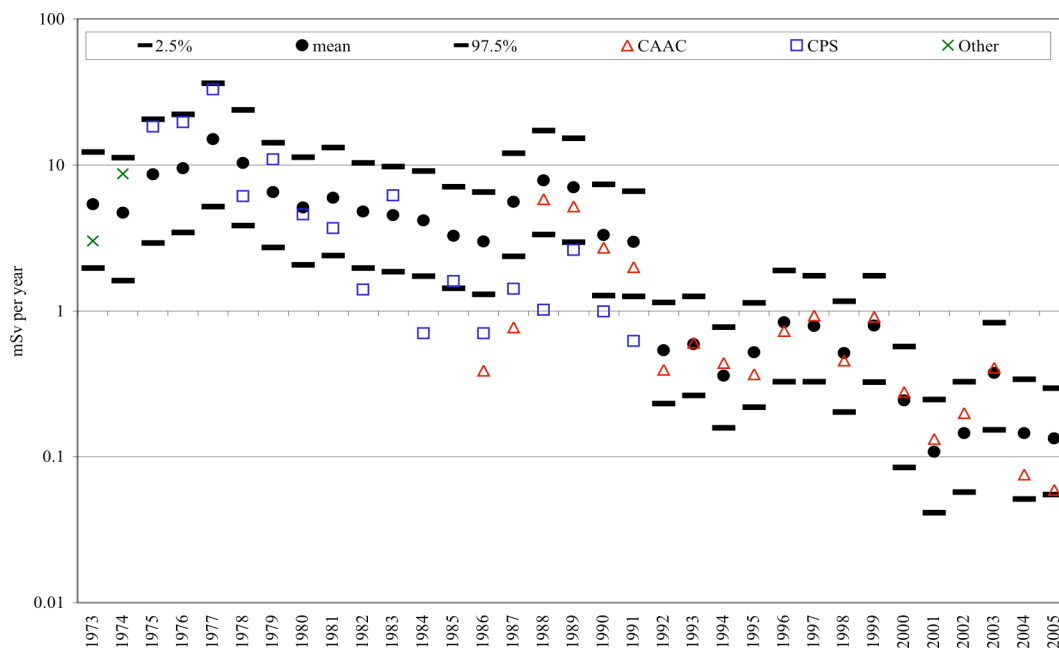


Figure 4. Annual mean doses with 95% confidence intervals predicted by DCART to the SW-MEI (Location Q, 1974 – 1978); Location VIS, 1973, 1979 – 2005) from release rates and dilution factors compared with historic tritium doses reported by LLNL. CAAC (Clean Air Act Code) represents AIRDOS-EPA and CAP88-PC; CPS is the Continuous Point Source Code.

Annual mean dose estimates calculated using DCART are higher than those reported by LLNL in 25 out of 39 comparisons; in twelve cases, mean doses predicted with DCART are higher than those reported by more than a factor of two. Ten reported doses fall outside the 95% confidence intervals of the DCART predictions: all are below the lower confidence limits. The agreement between dose predictions calculated by DCART and those of CAAC is very good between 1988 and 2003³³ when the ratio (DCART mean dose/reported CAAC dose) is 1.1, with a minimum at 0.73 (in 2002) and a maximum at 1.5 (in 1991). Agreement over the years between DCART and the Continuous Point Source (CPS) Code (Peterson et al. 1976), which calculated dose from inhalation only, is variable, although in later years the CPS doses consistently fall below the lower confidence limits of the doses predicted by DCART.

Accidental releases

Because ingestion dose may be much larger relative to inhalation dose after an acute release compared to a routine release of tritium, the assumptions behind the calculation of ingestion dose after an acute release must be carefully considered.

Predictions

Two sets of dose predictions and their 95% confidence intervals are shown in Figure 5. One set (“realistic”) is based on the assumption that the diet only contained those foods that would have been exposed to the tritiated plume at some point during their growth; the other set (“maximum”) is based on the assumption that the entire diet was contaminated except for grain. Because the UCL of the doses based on the realistic ingestion assumptions and the mean of the doses based on the assumption of maximum possible ingestion are similar, the two values were averaged to obtain what may be considered the highest likely dose. The UCL of the dose based on maximum ingestion is one, in all probability, that is far in excess of any dose that could have been received. It, however, may account conservatively for uncertainties that could not be quantified. The values for these two sets of doses are summarized in Table 5.

The highest dose predicted for an accidental release was 2,000 μSv (200 mrem) to a hypothetical MEI at Location R in 1954; 2,000 μSv was the UCL of the dose based on maximum ingestion assumptions. The highest likely dose for this release was about 360 μSv (36 mrem). The second highest predicted dose for an accidental release (i.e., the UCL of the dose based on maximum ingestion assumptions) was a factor of 2.4 lower than that for 1954. The highest likely dose in 1970 was 120 μSv (12 mrem), and the mean dose based on realistic ingestion assumptions (17 μSv [1.7 mrem]) was a factor of 3.8 lower than that for 1954. Uncertainty on both the 1954 and the 1970 dose predictions was high with factors of 1,200 and 4,900, respectively, between the UCLs based on the maximum assumptions and the lower confidence limits based on realistic ingestion assumptions. The third highest dose was predicted for the accidental release of 1965. The overall uncertainty for this release that accounted for the two sets of ingestion assumptions was a factor of 960.

³³ The 2004 and 2005 comparisons between DCART doses and LLNL reported doses are not consistent with earlier years because of a change to the ingestion assumptions for the dose reported by LLNL (see Appendix C in Part 3 of the TDR).

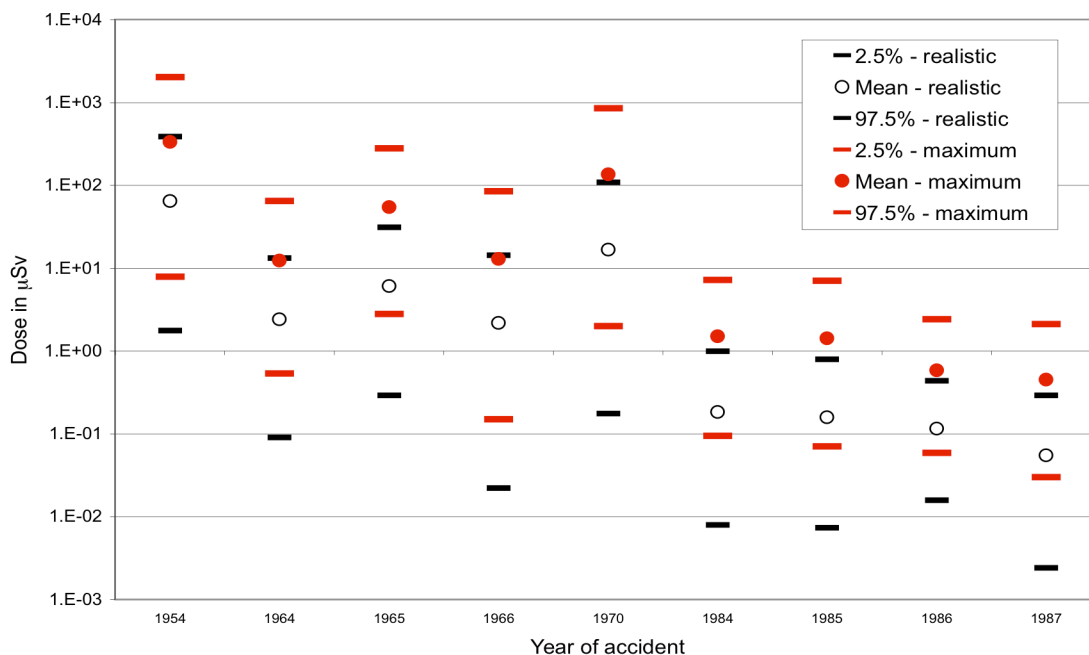


Figure 5. Predicted doses and their 95% confidence intervals for accidental releases from LLNL and SNL/CA. Realistic doses are based on the assumption that only a limited fraction of the diet could have been exposed to the plume; maximum doses assume all the diet was contaminated except grain.

Table 5. Highest likely and extremely conservative doses (μSv) to the MEI predicted for LLNL and SNL/CA accidental releases.

Source and year of release	Receptor location	Highest likely dose	Extremely conservative dose (UCL based on maximum assumptions)
LLNL 1954	R	360	2000
LLNL 1964	Q	13	64
LLNL 1965	X	43	280
LLNL 1966	Q	14	84
LLNL 1970	V	120	850
LLNL 1984	A	1.3	7.2
LLNL 1985	S	1.1	7.0
SNL/CA 1986	L	0.51	2.4
SNL/CA 1987	K	0.37	2.1

Mean dose impacts for 1964 and 1966 were essentially the same³⁴ to the same MEI (Location Q) and were only about 25% to 40% those for 1965³⁵ even though more than

³⁴ For 1964 the mean doses (based on maximum and realistic ingestion assumptions, respectively) were 12 μSv (1.2 mrem) and 2.4 μSv (0.24 mrem). For 1966 the mean doses (based on maximum and realistic ingestion assumptions, respectively) were 13 (1.3 mrem) μSv and 2.2 μSv (0.22 mrem).

³⁵ For 1965 the mean doses (based on maximum and realistic ingestion assumptions, respectively) were 54 μSv (5.4 mrem) and 6.1 μSv (0.61 mrem).

25 times more HT was released in 1965 than in the other two years and the UCL of the dose based on maximum assumptions was highest for 1965.

The doses and uncertainties for the accidental releases of 1984, and 1985, are within 10 - 15% of each other³⁶, even though the 1984 release was about 5,000 Ci HT and the 1985 release was only about 20% as large.

Dose predictions for the 1986 SNL/CA accidental release of HTO were somewhat higher than for the 1987 HT accidental release, but the uncertainty about the 1987 dose predictions was much greater than for 1986. The difference between the mean doses predicted using the assumptions about maximum and realistic ingestion ranges from a about factor of 5 for 1986³⁷ to about a factor of 8 for 1987³⁸.

The highest likely doses listed in Table 5 are undoubtedly conservative for some of the releases, and they are considerably lower than the UCLs of the doses based on maximum assumptions. The values (upper confidence limit of realistic dose followed by the mean of the dose based on maximum assumptions) from which the highest likely doses were calculated for the accidental releases with the greatest dose impact were

- 390 μSv (39 mrem) and 330 μSv (33 mrem) for 1954,
- 110 μSv (11 mrem) and 140 μSv (14 mrem) for 1970, and
- 31 μSv (3.1 mrem) and 54 μSv (5.4) for 1965.

The UCLs of predicted dose based on realistic assumptions and the mean of the distribution based on maximum assumptions for the accidental release with the lowest potential dose were 0.29 μSv (0.029 mrem) and 0.45 μSv (0.045 mrem) for the 1987 release.

Dose comparison for 1970

Doses from the 1970 accidental release were predicted both immediately after the release (Myers et al. 1973) and much later by the Agency for Toxic Substances and Disease Registry (ATSDR 2003) as part of a PHA of the Livermore site. Predictions calculated as part of this TDR for the accidental release of 1970 are compared with these earlier predictions in Table 6.

³⁶ For 1984 the mean doses (based on maximum and realistic ingestion assumptions, respectively) were 1.51 μSv (0.151 mrem) and 0.184 μSv (0.0184 mrem). For 1985 the mean doses (based on maximum and realistic ingestion assumptions, respectively) were 1.42 μSv (0.142 mrem) and 0.158 μSv (0.0158 mrem).

³⁷ From 0.12 μSv (0.012 mrem) for the mean dose based on realistic assumptions to 0.58 μSv (0.058 mrem) for the mean dose based on maximum assumptions.

³⁸ From 0.055 μSv (0.0055 mrem) for the mean dose based on realistic assumptions to 0.45 μSv (0.045 mrem) for the mean dose based on maximum assumptions.

Table 6. Comparison of doses (μSv) to the MEI predicted by the TDR, ATSDR (2003), and Myers et al. (1973) for the 1970 accidental release. Confidence intervals represent the 90th percentile (UCL is 95%). All doses, if rounded, were rounded to two significant figures so doses may not sum to the total. Symbols: A = ATSDR, T = TDR; M = Myers et al. T-m is the dose assuming a complete diet without grain; T-r is the dose assuming a realistic diet.

	Inhalation and skin absorption			Ingestion				Total Dose		
	A	T	M	A	T-m	T-r	M	A	T-m	T-r
Infant mean		0.96			190	22			190	23
Infant 95% CL		3.7			750	83			760	86
Child mean	340	1.4		4	200	22		340	200	24
Child 95% CL	1400	5.2		15	770	84	700	1400	780	89
Adult mean	100	1.1	30	1	130	16		100	140	17
Adult 95% CL	400	4.3		4	520	60		400	530	64

DISCUSSION

Predicted vs. observed concentrations of HTO in air moisture

Predicted HTO concentrations in air moisture are either indistinguishable from observed concentrations or they are higher. When observed and predicted distributions do not overlap, it is likely that the predictions are wrong. The incorrect predictions for the TDR, however, were always higher, and therefore any doses calculated from them would be guaranteed to be higher than any dose that could have been received. Reasons for such over-predictions were examined in Appendix B of Part 3 of the TDR.

Confidence intervals on the observed concentrations of tritium in air moisture (Figure 2) are greater than might be expected primarily due to the uncertainty associated with correcting for dilution of the sample by “bound water” in the silica gel (Guthrie et al. 2002; Peterson 2007a) and the uncertainty due to how well the sample represents the annual average (i.e., how many samples are missing or below the lower limit of detection). The uncertainty in recent years also includes high analytical uncertainty as the tritium concentrations in the samples have dropped. The highest uncertainty on the observations (a factor of 4.8) in 2005 was due to seven of the biweekly samples being below the detection limit.

Confidence intervals on the predicted concentrations of tritium in air moisture arise from uncertainty in dispersion, estimated release rates and the estimated mean annual absolute humidity. If observed and predicted concentrations of tritium in air volume were compared, there would be less uncertainty on the predictions (absolute humidity would not contribute) and slightly more uncertainty on the observations (due to uncertainties about the flow rate through the sampler).

The predicted fraction of HTO in air moisture derived from HT varied from year-to-year depending upon the fraction of total tritium released as HT. The fraction of HTO derived from HT for each year was small when it was assumed either that 54% of the annual release from the Tritium Facility was HTO or when the actual speciation of releases was used. The highest fraction when speciation was known peaked at about 6.6% in 1974 and 1975 when about 65% of the estimated total tritium released from the Livermore site

was HT. When it was assumed that the potential quantity of HTO released from the early Tritium Facility was only about 10% of the total, the predicted fraction of HTO in air moisture derived from HT was as high as 29% for 1957.

Although these results (Figure 2) and earlier ones (Peterson 2004) have demonstrated that CAP88-PC tends to overestimate air concentrations at VIS, it cannot be assumed that CAP88-PC overestimates air concentrations at the other potential locations of the SW-MEI. In the earlier test (Peterson 2004), when release rates from diffuse sources were lower than those assumed for the TDR (see discussion in Part 3 of the TDR), air concentrations at the ambient air tritium sampler, MESQ, located close to Location T, were underestimated during 1986 through 1992 by between a factor of 1.7 and a factor of 3.7 (median 2.2)³⁹. For CAP88-PC to correctly predict air concentrations⁴⁰ at potential locations of the SW-MEI, e.g., Locations T and Q, the assumption of area sources is necessary (whereas, for the Discovery Center, the overestimation of the dispersion model from stack sources makes the addition of area sources redundant or overly conservative).

Given the reasonable assumptions about potential unknown area sources in this TDR, given the large uncertainty associated with the early releases, and given the very conservative assumptions about diet, doses (which are directly proportional to predicted air concentrations) at Locations T or Q should not have been underestimated, although they would not have been overestimated by nearly as much as at the Discovery Center.

Annual dose predictions

The location of the SW-MEI is dependent upon the assumptions made about relative release rates, stack parameters, and the presence and location of assumed area sources. The effect on dose at any location from any one source cannot be predicted without dispersion modeling, and dispersion modeling must also be used to predict the location of the SW-MEI when release rates of sources change relative to each other. The SW-MEI when it was at Location T was very sensitive to the assumptions about stack heights, stack diameters and exit velocities used to determine the dilution factor for Building 231. A relatively small change in these assumptions would have shifted the location of the SW-MEI from Location T to Location S, at least for 1956, and from Location T to Location Q for 1960. This is not significant, however, because doses to the chosen SW-MEI and the other possible locations were within 10 – 40% of each other, well within the uncertainty of the calculations. Changing the assumptions about speciation only changed the ranking of the potential locations of the SW-MEI in minor ways.

A unit release rate (i.e., 1 Ci/y) from the Building 331 stacks produces a very small dose of about 1.2 μ Sv (0.12 mrem) to the SW-MEI when, for example, the SW-MEI is at Location VIS; the importance of the tritium releases from the Building 331 is due to their

³⁹ Compare this to the results for the VIS air tritium sampler, which, for the same time period, underestimated air concentrations in 1986 by 10%, and 1987 by 30%, and overestimated by 10 – 80% when *no* area sources were assumed; when a release rate for the Building 612 Yard was used that was half that assumed in the TDR, air concentrations at VIS were overestimated by up to a factor 2.3 and only underestimated by 4% for 1987.

⁴⁰ At LLNL, light winds blow to the west and northwest mainly at night when the stability classes are E and F. As a result, the highest air concentrations predicted by CAP88-PC from stack sources miss MESQ and come to ground about 3000 m from the source, well beyond MESQ. The plume has passed over MESQ and lack of vertical mixing with E, and especially F, stability class in CAP88-PC causes underestimation close to the source.

magnitude. The effects of releases from the Building 231 stack or Building 212 stack were higher per unit release at Locations T or Q than those of the Building 331 stacks because the release heights and exit velocities were lower and the facilities were relatively close to the receptor. The effect of an area source upon the receptor is roughly dependent upon the distance from a source to the receptor, but, given the wind and stability patterns of the Livermore site, there is a tendency for air concentrations to be somewhat higher at any set distance from a source to a receptor on the south and west sides of the site. Thus, for example, although the Building 331 WAA was slightly closer to Location VIS than it was to Location T, the estimated dilution factor at Location T was four times higher than that at Location VIS from this source.

There is considerable uncertainty surrounding the dose predictions for the earliest years (Figure 3). The predicted uncertainty becomes less after 1960 but, even though this TDR attempts to account quantitatively for all sources of uncertainty, the confidence intervals imply a more accurate dose assessment than is possible prior to 1973, at which time the molecular sieves were installed to sample for HT and HTO being exhausted from the Tritium Facility stacks. Among other things, it is impossible to quantify the uncertainty for sources that may or may not have existed in the chosen locations (e.g., the Building 231 WAA) or the uncertainty that arises when routine puff releases are modeled as steady-state and speciation is essentially unknown. However, because of deliberately conservative assumptions, reasonable confidence can be placed in the probability that any and all doses received by any member of the public will have been below the 97.5% confidence limit (based on 54% of the releases from the Tritium Facilities being HTO), no matter the location of the SW-MEI. Thus the UCL of the dose predictions shown in Figure 3 may be considered doses that could not have been exceeded.

The assumptions about the presence of area sources and the quantities of tritium released from them are intended to be conservative. The assumptions and their effect on the dose calculations may seem strange, however. For example, the assumption that the potential quantities of tritiated waste that had been stored at the Building 514 Yard were evaporated using the solar evaporation trays between 1962 and 1964 (the period during which solar evaporation was started and the Building 514 Yard had been converted to tanks but before the Building 612 Yard became operational) resulted in estimated release rates that were known to be absurdly high compared with what little was known about the quantities of tritium that were actually evaporated. The dose impact to the SW-MEI at the Discovery Center from this assumption was large because of the proximity (not known exactly) of the evaporation trays to the Discovery Center.

Ingestion dose is a larger fraction of the total annual dose for a release of HT than for a release of HTO of equal magnitude. In DCART, after an HT release, 89% of the adult's and child's doses comes from ingestion, and 93% of the infant's dose is obtained from food; in contrast, for a release of HTO, 80% of the total dose to adult and child is from ingestion, and 88% of the infant's dose is from food. This small difference occurs because, after a release of HT, soil microorganisms rapidly convert HT to HTO and the soil becomes a much more important source of HTO than it is after a release of HTO. As a result, the concentration of HTO in TFWT after a release of HT is higher than the concentration of HTO in air moisture, while, for a release of HTO, the concentration in the TFWT is lower than that in air moisture because the soil is a relatively unimportant

source of HTO compared with the air⁴¹. This effect is most dramatic for root crops, which are assumed to have a concentration approximately equal to the concentration in soil water. After a release of HT, it is estimated in DCART that the dietary contribution to ingestion dose from root crops is double that seen after a release of HTO.

A sensitivity analysis demonstrated that, although the distributed HTO dose coefficient used in DCART has an uncertainty less than that of the OBT dose coefficient, dose is more sensitive to it because so much of the dose is derived from ingested HTO.

The UCLs for doses from releases between 1953 and 1960 (Figure 3) are comparable to the UCLs for doses predicted between 1961 and 1973 (the years when stack sampling occurred and releases were reported quarterly rather than as occurring on a specific date) because all were calculated assuming that 54% of the releases from either Building 231 or Building 331 were HTO. The magnitude of the confidence intervals on doses between 1953 through 1960 is much larger than in later years because it is the result of combining the confidence intervals of two sets of dose calculations with different assumptions in an attempt to embrace the full potential uncertainty: the 2.5% confidence limit and mean were calculated based on the assumption that most of the tritium released was HT due to the known nature of the releases as puffs occurring a very few times a year.

From 1961 until 1974 (when releases from the Tritium Facility were measured as HT and HTO), the confidence intervals are based on the one dose calculation that assumed that 54% of the tritium released from the Building 331 was HTO. In reality, the shift from releases of mostly HT to 54% HTO (a ratio based on monitoring and speciation data for normal operations) undoubtedly was gradual and did not change abruptly, as it did for the TDR modeling for 1961. The real rate-of-change of the speciation for the releases between 1962 and 1973 is unknown, of course, and, because the upper confidence limit will be representative or conservative when 54% of the release is assumed HTO⁴², there was no incentive to attempt to model a gradual, and unknown, increase in the fraction of HTO released over time. The means and 2.5% confidence limits for 1961 through 1973 may in reality be lower than shown in Figure 3 because of the probability that the fraction of HTO in the releases from Building 331 was less than 54%, at least for a few years after 1961. The mean (or best estimate) of the distributions for all years may be higher or lower than their positions within the confidence intervals depending upon the unknown relationship between actual and assumed speciation of releases and on the presence of area sources.

From 1974 onwards, the release rates of both HT and HTO from Building 331 were measured, and the measured values⁴³ were used to calculate the dose distributions.

Location T was the SW-MEI when releases were primarily from Building 231 and its (assumed) waste accumulation area. Doses in 1957 were higher than other years because about 12,000 Ci (444 TBq) were released that year (Southwick 1957); more than 11,000 Ci of HT were released from Building 231 (the upper confidence limits were calculated

⁴¹ See equation 4 in Part 1 of the TDR.

⁴² Uncertainty on speciation was included with the uncertainties on the release rate and ranged from $\pm 18\%$ to $\pm 40\%$ depending upon whether release was HT or HTO and from which stack the tritium was emitted.

⁴³ The fraction of the total tritium that was released as HTO from each stack varied between 0.05 (Stack 1 in 2004) and 0.97 (Stack 2 in 2005).

on the assumption that 6,480 Ci [240 TBq] of the total tritium released were HTO). The UCL is especially high because of the uncertainty on the release rate due to one of the quarterly reports not having been found. The mean (assuming 5.4% HTO) is about ten times lower than the UCL (assuming 54% HTO). As soon as Building 331 started to release tritium, because of the magnitude of releases and the location of Building 331 relative to Location T and Location VIS, the SW-MEI shifted to VIS for all years except 1961 and 1974 – 1978, when the SW-MEI was at Location Q.

Dose predictions based on observed concentrations of HTO in air include the contribution of HT that was converted to HTO in the environment, but, because there is no way to distinguish HTO converted from HT from that released as HTO, there is no way to determine from observed air concentrations the fraction of dose contributed by the HTO that arose from converted HT. A model is needed for this calculation. In DCART, a release of HT has about a 20% greater effect on dose than it does on HTO concentrations in air for the same reasons (described above) that result in ingestion dose being relatively higher after a release of HT compared with after a release of HTO.

That the entire diet could have been contaminated or that a person stayed home 24 hours a day for the entire year, as is assumed in DCART, is highly unlikely. Dose predictions for two years with high doses were recalculated using two different sets of assumptions⁴⁴ to reduce the portion of the diet that could have been contaminated and to reduce the hours of occupancy at the location of the SW-MEI. For both approaches, it was assumed that no grain could have been contaminated (grain, although historically grown in the Livermore Valley, was not grown as close to the Laboratory as the locations of the SW-MEI and was most certainly not grown in the garden of the hypothetical SW-MEI) and the inhalation rate was adjusted to reflect occupancy of the SW-MEI with a uniform distribution of 0.5 – 0.9 (50% – 90%)⁴⁵.

In one set of calculations, the various kinds of food ingested were assumed, with equal probability, to be uncontaminated (i.e., not grown locally), contaminated (i.e., the default in DCART), or some fraction of uncontaminated and contaminated. Thus the parameter (“fraction of the consumed food arising from the contaminated source”) in DCART was changed from a deterministic value of 1 to a uniform distribution (0 – 1) for each foodstuff. In the other set of calculations, the fraction of contaminated food was defined by a triangular distribution of 0 – X – 1, where “X” was the mean fraction of home-grown foodstuff in the Western United States from the U.S. Environmental Protection Agency (EPA) “Exposure Factors Handbook (US EPA1999)”⁴⁶.

Using the first set of revised assumptions, the mean doses to an adult, child, and infant were reduced to about 40% of the mean doses obtained using a completely contaminated diet; dose to an infant was reduced to about 45%. The 97.5% confidence limit was reduced slightly less – about a factor of two. Using the second set of revised assumptions, the predicted mean doses to an adult, child, and infant were reduced by a

⁴⁴ Although doses were calculated for 1957 using one set of assumptions and for 1977 using the other set, the conclusions affect all years more or less equally.

⁴⁵ Lognormal distributions for inhalation rates representing the occupancy factor were calculated using Crystal Ball® to be $3,400 \pm 1,250 \text{ m}^3 \text{ y}^{-1}$ for the adult, $3,450 \pm 1,600 \text{ m}^3 \text{ y}^{-1}$ for the child, and $1,150 \pm 822 \text{ m}^3 \text{ y}^{-1}$ for the infant.

⁴⁶ “X” equals 0.015 for leafy vegetables, 0.084 for root crops, 0.12 for fruits and non-leafy vegetables, 0.007 for dairy, 0.041 for beef, 0.011 for pork, 0.008 for poultry and 0.021 for eggs.

factor of three compared with the mean doses obtained using a completely contaminated diet. The 97.5% confidence limit was reduced slightly less by about a factor of 2.5. Thus, assuming an intake of contaminated food that is still conservative (because all foods except grain are potentially contaminated to some extent) but not excessively so, the total annual dose from a routine release of tritium can be reduced by a factor of two to three.

At Location T, the most conservative assumptions resulted in a maximum predicted dose with an UCL to an adult for 1957 of 370 μSv (37 mrem) (see Figure 3) and 590 μSv (59 mrem) to an infant. In 1958 (NCRP 1958), the National Council on Radiation Protection and Measurements (NCRP) recommended a dose limit for continuous exposure of 500 mrem (5 mSv) to “persons outside of controlled areas”. The hypothetical dose to an infant from LLNL operations was just 12% of this dose limit. In fact, this maximum dose received by a hypothetical infant using conservative assumptions in 1957 is below the current DOE radiation standard for protection of the public of 1 mSv y^{-1} (100 mrem y^{-1}) effective dose equivalent for prolonged exposure of a maximally exposed individual in an uncontrolled area (DOE 1993) for all types of releases. Dose at Location VIS never exceeded the NESHAPs limit of 100 μSv (10 mrem) annually regardless of the degree of conservatism in the predictions.

LLNL historical annual dose predictions compared with those of the TDR

Given the very different assumptions of DCART compared with the models used by LLNL to calculate reported annual doses to a member of the public, the agreement between doses predicted by DCART and those reported historically by LLNL is remarkable. However, it is useful to understand the various compensatory factors that caused the resulting agreement (see also Appendix C in Part 3 of the TDR).

Starting in 1973, LLNL reported annual tritium doses to the public. That year the dose was calculated from the highest weekly observed air tritium concentration obtained from the newly established air tritium monitoring network. From 1974 on, dose was based on dispersion modeling. Up until 1985, the dose from each facility was reported as “fence post” dose, which is the equivalent of the currently used MEI, and the doses from each facility were not summed in the SAERs⁴⁷. Although the summation of these doses, each of which was calculated for a different location, would have been higher than dose to the SW-MEI (had such a dose been calculated), it is the only value that can reasonably be compared with dose to the SW-MEI predicted for the TDR (Figure 4). From 1986 through 1991, the doses were reported for each facility at a perimeter location (unspecified). These doses were added together in the SAERs, which implies that dose to a SW-MEI was being calculated.

From 1992 onwards, LLNL, for compliance with NESHAPs, reported doses to the MEI and to the SW-MEI from both monitored stacks and diffuse area sources. The SW-MEI

⁴⁷ The fence post dose, although not always identified, was most probably along the Laboratory’s southern boundary with East Avenue for the Tritium Facility and Building 212, at least. As demonstrated in the TDR for 1973 through 2005, the location of the SW-MEI was along East Avenue from 1974 through 1978 (observed concentrations at the air tritium sampling location CAFÉ [adjacent to East Avenue] were about 50% greater than concentrations at VIS during the early years of air sampling).

was defined at that time as the UNCLE Credit Union (Surano et al. 1993). As a result, the most similar, or at least less variable, comparison between DCART predictions and doses reported by LLNL occurred after 1992. For these years, inhalation and ingestion doses were predicted for similar locations (the Discovery Center is very close to the UNCLE Credit Union). Although inhalation and ingestion assumptions in DCART and CAP88-PC are very different and the dose coefficients are different, when HTO concentrations in air and absolute humidity are the same, when neither model includes any contaminated drinking water, and when both models assume all food is homegrown, DCART's adult dose is 43%⁴⁸ that predicted by CAP88-PC (for 1992 through 2003). In 2004, when LLNL changed the ingestion assumptions in CAP88-PC to be more realistic (yet still conservative), DCART's adult dose became 110%⁴⁸ of the comparable dose predicted using CAP88-PC. This change is reflected in the comparisons shown in Figure 4 where CAP88-PC predictions, which were approximately the same as DCART's mean predictions for many years, suddenly drop close to the 2.5% confidence limits of the DCART predictions.

There are other factors that account for differences between the doses reported by LLNL and those predicted from ambient (see Figure 6 in Part 3 of the TDR) or predicted air concentrations (see Figure 8 in Part 3 of the TDR) using DCART. For instance, prior to 1992, only doses from stack releases were reported by LLNL; from 1992 onwards, doses from both stack and diffuse sources were reported. Also, between 1973 and 1988, only tritium inhalation doses to the MEI or SW-MEI from stack releases were calculated using dispersion modeling⁴⁹. Inhalation has always been the most important pathway to tritium dose for workers, and that perspective probably was applied when calculating dose to the public. With the advent of the CAAC, it was recognized that ingestion dose from tritium has the potential to be much more important to a member of the public than dose from tritium inhalation. (In DCART and CAP88-PC, inhalation only contributes about 20% or 16%, respectively, of a total tritium dose that assumes all food but no drinking water is contaminated.) In addition, for some years, LLNL modeled all releases as HTO, while in other years, LLNL calculated dose from releases of HTO only.

Some differences between doses reported by LLNL and those predicted by DCART will be due to dispersion modeling. Gaussian models may differ in the way in which σ_y ⁵⁰ and σ_z ⁵¹ are calculated, the way plume rise is calculated, whether or not, or how, building wake effects are handled, and the ways in which deposition and plume depletion are taken into account. Differences such as these in the models will result in the prediction of different dilution factors. The quality of the meteorological data collected and the way the data are formatted as input to the model can also affect predicted dilution factors. The result of these differences is that each Gaussian dispersion model will calculate a different dilution factor for each different set of meteorological data. In the case of

⁴⁸ Note that this relationship (which is not obvious in Figure 4) is based on deterministic parameters in DCART. DCART's deterministic doses are always lower than its stochastic doses because of the uncertainty distributions on the dose coefficients (Peterson 2006).

⁴⁹ Ingestion doses were calculated in the SAERs from 1979 onwards using equations from the US Nuclear Regulatory Commission's Regulatory Guide 1.109 (US NRC 1977) and observed concentrations in sampled vegetation or foodstuff, e.g., milk; these doses were reported along with the measured environmental concentrations of the vegetation or foodstuff in data tables.

⁵⁰ Standard deviation of the plume width in a horizontal direction

⁵¹ Standard deviation of the plume width in a vertical direction

CAP88-PC, there is only about $\pm 10\%$ difference between dilution factors calculated from annual or four- or five-year wind files for each of the historical tritium sources with VIS as the receptor. This small difference is due largely to the fact that the wind blows towards VIS from the tritium sources with high frequency (about 54% of the time winds blow from the south-southwest through west directions). In contrast, the wind blows towards Location Q from most major sources with a much lower frequency than it does towards the Discovery Center. As a result, the variability of dilution factors for a single facility derived from different wind files used as input to CAP88-PC for Location Q can be as much as $\pm 40\%$.

Between 1979 and 1995, the SNL/CA TRL released HT and HTO to the atmosphere. The dose contribution of SNL/CA should not be included as part of LLNL's dose impact, but it has been included in the TDR so that a direct comparison between doses predicted from ambient air concentrations and from dispersion modeling can be made. The percentage of total dose at the Discovery Center predicted to have been contributed by the TRL ranges from 1.9% (1979 – 1984) to 30% (1992 – 1994); the mean contribution for the years not listed is 14%. The impact of SNL/CA releases at VIS is not obvious (Figure 4) even for those years when they contributed a calculated 30% of the dose (compare the predictions of LLNL, which did not account for SNL/CA releases, with the means of the dose distributions predicted by DCART, that did).

Predictions for accidental releases

The challenges faced in determining reasonable yet conservative dose predictions for historical accidental releases are similar, but not identical, to those for historical routine releases. As long as meteorological data used in the dispersion model was collected for a particular year or years and represented local conditions, the routine doses predicted for other years using that wind file should be reasonably certain. In contrast, even when 15-minute meteorological data are used in a dispersion model, the uncertainty about the dilution factor is greater; when meteorological data at the time of the accidental release are unknown, the daunting challenge is to select conditions that will result in a dose that is not underestimated but is also not the worst case with a very small probability of occurrence.

Ingestion dose is potentially much more important after an acute release, particularly of HT, than from a routine release. In DCART, the ingestion/inhalation ratios for dose to an adult for HT and HTO are factors of about 8 and 4, respectively. The ingestion/inhalation ratio derived from UFOTRI has a uniform distribution of 80 – 200 for HT and a uniform distribution of about 4 – 20 for HTO (this varies with what is known about the meteorology). When the dose is being calculated for an entire year, it may be reasonable to assume the entire diet was contaminated, because all plants that were growing at any time during the year would have been exposed to the tritium in air. Depending upon the time of year when the accidental release occurred, however, the portion of the diet that is potentially contaminated can be very different. To assume that the entire diet is contaminated after an acute release is overly conservative.

Dispersion modeling for accidental releases and air concentrations

The magnitude of the dilution factor for an acute release depends upon how close the MEI is to the source, upon stack parameters, and upon the meteorological conditions during the release.

Given the assumptions of the TDR, the minimum uncertainty about the dilution factor was a factor of 16 (UCL divided by lower confidence limit). This factor of 16 was the uncertainty about the dilution factor for the years when the meteorological data were known (1985, 1986, and 1987); for June 1984, when the maximum likely dilution factor from the 15-minute data resulted in an uncertainty less than that assigned ($\pm 80\%$); and for 1964, when the use of hourly mean data suppressed the variation that results in potential maximum dilution factors when 15-minute data are used. The highest uncertainties were for 1966 (a factor of 196 because of the maximum potential dilution factor derived from the generic 15-minute data) and 1970 (a factor of 167 because of the large variation in dilution factors at Location X due to the extremely variable winds).

During a normal workday from about 8 am until 5 pm, stability classes are usually restricted to A, B, C, and D, particularly in the summer. Under these conditions (e.g., June 1984), there is not a great deal of variability in the dilution factors. However, because it is known that the 1970 release occurred at 6:00 am and because operations are known to have occurred outside core work hours, meteorological conditions that account for early morning and late afternoon (i.e., include lower wind speeds and stability classes E and F), at the very least, must be included when estimating the dilution factors for the accidental releases in 1954 and 1966⁵² that occurred at unknown times. The potential dilution factors will be higher when early morning and early evening meteorological conditions are taken into account, and the uncertainty about the dilution factors may be much greater than when the time of release is known.

Another unknown for the releases of 1964 and 1966 was their duration. The major releases of 1965 and 1970 were less than 10 minutes and 30 minutes respectively. The shorter the release, the higher the dose impact at any one particular location, although if conditions are stable, the duration of the release will have less effect on the time-integrated air concentration than if conditions are unstable. Although some accidental releases extended over several hours (e.g., the accidental releases of 1984 and 1985 lasted 2.5 and nearly 7 hours, respectively), nearly all of the tritium in those incidents was released in the first 30 minutes. Thus, to err on the side of conservatism, no release of unknown duration was assumed to have exceeded 30 minutes.

In addition to using HOTSPOT to estimate the dilution factors for the LLNL and SNL/CA accidental releases, air concentrations predicted by HOTSPOT were compared with air concentrations predicted by ATSDR (2003) for the 1965 and 1970 releases and with those predicted at the time of the release in 1965 (Peterson et al. 2002). The predictions using HOTSPOT were made at the same distances using the same wind speeds and stability classes as were used in the other modeling efforts. The time-integrated concentrations of HT in air predicted by HOTSPOT at the location of the ATSDR MEI were within 30% of ATSDR's results for 1965 and within 2% for 1970.

⁵² The time of release was not known in 1964 either, but, as mentioned, the use of hourly data damped the potential for high dilution factors.

The air concentrations at distances between 0.1 and 100 km predicted by HOTSPOT averaged 20% higher than the predictions made at the time of the 1965 release. This level of agreement between model predictions is excellent.

Dose predictions for accidental releases

Of the two sets of dose predictions that were calculated for all accidental releases (Figure 5), the confidence interval for the dose based on realistic ingestion assumptions is where the most likely dose from each accidental release is expected to have fallen. For those releases with known meteorological data, the UCL of the dose calculated from realistic ingestion assumptions, when averaged with the mean of the dose calculated from maximum ingestion assumptions, may be considered, because of some conservative assumptions, a dose that could not have been exceeded. The potential dose from releases with known meteorology could be as low as the lower confidence limit on the realistic dose.

For those releases when either the meteorological conditions were unknown (1954, 1964, 1966, and 1984) or when the conditions of release were not well understood (1986), the UCL of the dose based on maximum ingestion assumptions has been assumed to be the dose that could not have been exceeded. This is considered a prudent approach when it was impossible to make a true assessment of uncertainty. The dose predicted *at the location of the MEI* will fall between the UCL based on the maximum ingestion assumptions and the lower confidence limit based on the realistic assumptions. If the receptor lived at the location selected for the MEI, the UCL of the dose based on maximum ingestion assumptions is the dose that could not have been exceeded, but, because the location of the MEI for these releases was chosen as the closest structure to the release, the dose to an actual receptor could have fallen below the lower 2.5% confidence limit. For example, the probability that the daytime winds would have been blowing over the closest possible structure to the source (Location R) at the time of the release in October 1954 was less than 10%; in April 1966, the probability that the plume would have passed over Location Q at the time of the release is smaller – less than 5% (estimates based on Gouveia and Chapman 1989). Thus the probability is very good that the dose received by the actual MEI was less than that calculated in the TDR because the wind most probably was blowing towards a receptor much farther from the source than was the conservatively chosen MEI⁵³.

The dose that could not have been exceeded (2,000 μSv [200 mrem]) predicted to a hypothetical member of the public for the 1954 accidental release of HTO was the highest dose predicted in the TDR because HTO (rather than HT) was released, the meteorology was not known so that conservative assumptions about ingestion were made to compensate for any potential failure to account for all the uncertainty on the dilution factor, and the closest public structure to the release point was assumed to be the location of the MEI. The magnitude of the dose received by an actual MEI is most likely considerably overestimated, given the small likelihood that the winds were blowing towards Location R at the time of the release. If it could be said with certitude that the

⁵³ On average, the concentration (or dose) for a radionuclide that doesn't deposit or that gets re-emitted quickly, like tritium, will drop off with distance raised to the power 1.2. For other radionuclides, or for tritium in this TDR when HOTSPOT was used without the deposition of tritium, the concentration is expected to drop off with distance raised to the power 1.5 (UNSCEAR 2000).

uncertainty applied to the dilution factor were accurate, then, even with the wind blowing directly over Location R, the dose that could not have been exceeded would have been the same as the highest likely dose (360 μSv [3.6 mrem]).

The predicted dose impact for 1970 is, although highly uncertain, as accurate as it can be, given the methods of predicting dose and the assumptions made. The dose to an adult that could not have been exceeded was 120 μSv (12 mrem).

Dose from the 1970 release was also calculated immediately after the release (Myers et al. 1973) and as part the ATSDR's PHA (ATSDR 2003) (Table 6). The locations of the MEI for the 1970 release were different for the ATSDR, Myers et al., and TDR assessments. For Myers et al., the inhalation dose was calculated at the nearest perimeter location downwind of the release (0.6 to 0.75 miles [0.96 – 1.2 km]); ATSDR calculated the inhalation dose at the location of the maximum air concentration (1 -1.5 miles [1.6 – 2.4 km]); inhalation dose for the TDR was calculated at the location of the closest structure downwind of the Livermore site (2.0 km). The Myers et al. dose (30 μSv [3.0 mrem]) was based on the conservative assumption that 1% of the release was HTO; ATSDR's mean dose to an adult (100 μSv [10 mrem]) was based on the inhalation of HTO emitted from the soil for the 12 days following the release. The result for the TDR (1.1 μSv [0.11 mrem]) is much lower than even the prediction of Myers et al. because only the upper end of the HTO/HT inhalation dose ratio based on UFOTRI modeling equals the assumption that 1% of the release was HTO and because the MEI for the TDR was about twice as far away as that for the Myers et al. calculation (which would mean roughly a decrease in dose by a factor of 2.3 from 30 μSv [3.0 mrem] to 13 μSv [1.3 mrem]). Using the dispersion assumptions of the TDR in HOTSPOT and the assumption of Myers et al. that the release was 1% HTO, an inhalation centerline dose of 21 μSv at 1.2 km was calculated; when a wind speed of 1 m s^{-1} was used (the low end of the range of wind speeds reported in Myers et al. [1973]), the inhalation centerline dose predicted by HOTSPOT rose to 29 μSv (2.9 mrem) at 1.2 km. The dispersion modeling and inhalation assumptions used by Myers et al. are confirmed by HOTSPOT.

Using the UFOTRI equation method and assuming the release had been 1% HTO, the mean tritium dose from inhalation and ingestion (regardless of ingestion assumptions) was calculated to be between 20 and 34% higher for adult, child, and infant than it was when the release was assumed to be 100% HT. The dose that could not have been exceeded assuming the release was 1% HTO was 140 μSv (14 mrem) to an adult and 210 μSv (21 mrem) to a child.

The ingestion dose to a child (700 μSv [70 mrem]) calculated by Myers et al. was intended to be impossibly high. It was based on the assumption that the child drank milk from a cow (assumed, contrary to fact, to be onsite) that consumed the grass that had the highest measured concentration of tritium in it. The Myers et al. estimate and the 95% confidence limit for the dose predicted by the TDR for maximum ingestion assumptions are very similar (i.e., also impossibly high). The ATSDR 95% confidence limit for ingestion dose to a child is lower by about a factor of 50 than either the Myers et al. or TDR predictions based on maximum ingestion assumptions. ATSDR used the concentrations measured in vegetation and milk offsite after the accident, along with the water contents and a factor to account for the dose effect due to OBT, to calculate their ingestion doses. However, ATSDR's approach did not account for the build-up of OBT

in the plants from the HTO emitted from the soil during the days following the release. Nevertheless, ATSDR's predicted ingestion doses to child and adult fall within the TDR's 90%⁵⁴ confidence intervals (the 5%⁵⁴ confidence limits for the child and adult in the TDR are 0.41 and 0.28 respectively) for the doses based on realistic assumptions. The uncertainty on the TDR's ingestion doses also embraces the predictions of Myers et al.

The total doses to child and adult predicted by ATSDR and this TDR (dose based on maximum ingestion assumptions) are similar, but the assumptions behind them are very different, as described above. The dose assumptions demonstrate clearly that, as ATSDR affirmed based on similar or even larger doses, these doses were well below levels of public health concern.

The 1965 HT release was the largest in LLNL history. Because there is a record of the meteorological conditions during the release and the nearest possible location for the MEI was nearly 2 km from the release, the mean dose consequences of the 1965 release were about 40% that of the HT release of 1970 and 14% of the HTO release in 1954. The dose that could not have been exceeded was 43 μSv (4.3 mrem). ATSDR (2003) did not explicitly calculate doses from the 1965 release because they determined that at the location of maximum impact to the public, air concentrations and deposition were less than for the 1970 release. ATSDR reasoned that if there was no public health hazard from the 1970 release, then there would be none from the 1965 release. A mean dose to an adult in 1965 of 9 μSv (0.9 mrem) can be estimated based on soil emission rates predicted by ATSDR scaled to the difference in magnitude between the 1965 and 1970 accidental releases. This estimate of 9 μSv (0.9 mrem) is higher than the mean of 6.1 μSv (0.61 mrem) based on realistic ingestion assumptions predicted for the TDR.

Mean dose impacts for 1964 and 1966 were essentially the same⁵⁵ because the magnitudes of the release were assumed similar and because the dilution factors for 1964 and 1966 were essentially identical. The predicted doses for these releases were relatively large compared with the dose from the release in 1965. This can be explained because the dilution factor for 1965 was about nine times smaller than the ones for 1964 and 1966 and the MEI was at Location X (distant from the source) rather than at Location Q (close to the source). The doses that could not have been exceeded were 64 μSv (6.4 mrem), 43 μSv (4.3 mrem), and 84 μSv (8.4 mrem) for 1964, 1965, and 1966 respectively. The doses for 1964 and 1966 that could not have been exceeded were the UCLs of the dose predictions based on maximum ingestion assumptions because the meteorology was unknown; for 1965, the dose that could not have been exceeded was the same as the highest likely dose (Table 5) because the meteorology during the release was known.

The uncertainty about the dose predictions for 1964 (the UCL of the maximum dose divided by the lower confidence limit of the dose based on realistic assumptions) is a factor of 710, which is smaller than the confidence intervals for the other two accidental releases without known meteorological conditions (1954 – a factor of 1,200 and 1966 – a

⁵⁴ This level of probability was selected instead of the normal 95% confidence interval used for the TDR because a 90% confidence interval was used by ATSDR for calculations.

⁵⁵ For 1964 the mean doses (maximum and realistic, respectively) were 12 μSv (1.2 mrem) and 2.4 μSv (0.24 mrem). For 1966 the mean doses (maximum and realistic, respectively) were 13 (1.3 mrem) μSv and 2.2 μSv (0.22 mrem).

factor of 3,800). This relatively small confidence interval is counter-intuitive, because the 1964 accident was the one that might not have occurred and had the largest uncertainty about the source term. The size of the 1964 confidence interval is small, however, because the meteorological data used to select the extreme conditions to obtain the highest likely dilution factors were obtained from hourly averages in which the fluctuations seen in the 15-minute data are damped out. Thus the uncertainty about the dose predictions may be artificially small. Applying the greatest magnitude of uncertainty seen in the TDR (that for 1970), the 1964 dose could be shifted upwards by about a factor of seven. The confidence interval on the 1966 release is large because of the generic meteorological data for April.

Doses from the accidental releases of 1984 and 1985 were an order of magnitude or more lower than doses from earlier accidental releases. The similarities between these doses, in spite of the difference in the source terms, were due entirely to the difference in the dilution factors. Note, however, that because of inherent uncertainty that arose from not having meteorological data for 1984, to be health protective, the dose selected as the one that could not have been exceeded was the UCL of the dose predicted from maximum ingestion assumptions (7.2 μSv [0.72 mrem]) in contrast to the dose that could not have been exceeded in 1985 – when the meteorology at the time of the release was known – of 1.1 μSv (0.11 mrem), which was the highest likely dose (Table 5). Compared this way, the doses bear some resemblance to the quantities of HT released (5,500 Ci in 1984 compared with 972 Ci in 1985).

Although the UCLs of the doses based on maximum ingestion assumptions for the two SNL/CA accidental releases in 1986 and 1987 are within about 10% of each other⁵⁶, the mean dose based on realistic assumptions for 1986 is twice that of 1987⁵⁷. On the one hand, the accidental releases were very different (HTO in 1986 and HT in 1987; duration – 480 – 600 minutes in 1986; 2 minutes in 1987). On the other hand, the meteorological conditions were similar (D stability and about 4.5 m s⁻¹ mean wind speed). The MEI in both cases was more than 1 km from the stack. The calculated uncertainty about the HT release was greater than for the HTO release due to the additional uncertainty associated with converting HTO inhalation doses predicted by DCART to HT inhalation doses before the ingestion to inhalation ratio was applied. The dose that could not have been exceeded for this HT release in 1987 was 0.37 μSv (0.037 mrem). Because the true uncertainty about the 1986 HTO release is unknown and potentially large, even though meteorological conditions were well characterized, the dose that could not have been exceeded of 2.4 μSv (0.24 mrem) was based on the UCL of the dose based on maximum ingestion assumptions.

The releases of 1985, 1986, and 1987 were small. They would not have been modeled as accidental had they occurred in years of higher routine release rates.

The dose impact of a release of HTO is, as expected, considerably greater than that of a release of HT. The dose ratios (HT dose/HTO dose) calculated by UFOTRI for the same release rates for the three potential locations for ITER ranged from 2.9 to 6.6% (Raskob et al. 1999). For the hypothetical HT and HTO scenarios of EMRAS, UFOTRI's dose

⁵⁶ 2.4 μSv (0.24 mrem) in 1986 and 2.1 μSv (0.21 mrem) in 1987.

⁵⁷ 0.12 μSv (0.012 mrem) for 1986 and 0.055 μSv (0.0055 mrem) for 1987.

ratios ranged from 0.8 to 4%. The dose ratios are very sensitive to the meteorology assumed but are unlikely to be much higher than about 7%.

CONCLUSIONS

In general, the goal of a dose reconstruction is to estimate, to the extent possible, actual exposures and doses received by real people. This TDR has not done that. Rather, conservative assumptions were made to assure that the dose was not underestimated. Although all doses were modeled using a probabilistic approach, with realistic distributions for most input parameter values, certain assumptions were intended to guarantee health protective over-predictions.

- All structures open to the public (such as the automotive garage [Location Q] or the Discovery Center [Location VIS]) were potential residences of the SW-MEI or MEI.
- All food grown was exposed to a single air concentration calculated for a single cubic meter of air.
- The SW-MEI or MEI lived at the location 100% of the time.
- The assumed ingestion rate of homegrown foods was much greater than expected for an individual growing a few vegetables for home consumption. When annual doses from routine releases were predicted, the entire diet was assumed equally contaminated.
- For accidental releases, the highest likely dilution factor was included in the distribution when meteorological conditions were unknown.
- For accidental releases, the location of the MEI was the nearest structure to the release point when meteorological conditions were unknown.
- After an accidental release, ingestion of vegetables could have occurred within in an hour of the passage of the plume.
- Additional conservatism was built into the dose predictions at Location VIS, because air concentrations are over-predicted by CAP88-PC at that location.

Because of the assumptions, calculated air concentrations and resulting doses predicted at the location of the SW-MEI or MEI were higher than they could have been at any actual residence⁵⁸. Exposure to routine or accidental releases of LLNL (and SNL/CA) tritium by real people at real residences would have been lower because air concentration is more or less inversely proportional to the square of the distance from the source, and most of the diet would not have been homegrown.

Based on these assumptions, a dose that could not have been exceeded was defined. For routine releases, this was the UCL of predicted doses based on the assumption that 54% of the tritium released from the Tritium Facility was HTO (when speciation was unknown), based on observed release rates of HT or HTO (when speciation was known),

⁵⁸ Even the dose at Location VIS that was calculated from observed concentrations of tritium in air would have been a greater dose than could have been received by any actual member of the public.

or, when the SW-MEI was at the Discovery Center during the years of ambient air tritium monitoring, based on the concentrations of HTO in air measured at the VIS air tritium monitor. For accidental releases, the dose that could not have been exceeded was the mean of the UCL of doses based on realistic ingestion assumptions and the dose based on maximum ingestion assumptions (when meteorological conditions during the release were known) or the UCL of doses based on the maximum ingestion assumptions (when meteorological conditions during the release were unknown). For some of the accidental releases, the dose received by an actual individual might have been less than the lower confidence limit of the dose distribution based on realistic ingestion assumptions.

Annual doses from routine releases

To calculate dose from routine releases, CAP88-PC was used to obtain dilution factors for each facility at the locations of the SW-MEI. These dilution factors were then used with release rates for all facilities in DCART to predict doses at the three locations of the SW-MEI, Locations T (1953 – 1958), Q (1961, 1974 – 1978), and VIS (1959, 1960, 1962 – 1973 and 1979 – 2005). Concentrations in air moisture at Location VIS were also predicted to compare with annual mean air moisture concentrations measured at the VIS air tritium sampling unit between 1973 and 2005.

Predicted air moisture concentrations at the VIS air tritium monitor were usually within a factor of two of the observed mean annual concentrations, and the confidence intervals on the predictions overlapped the confidence intervals on the observations for all years except when the predicted air moisture concentrations exceeded observed air moisture concentrations by more than a factor of two. The reasons for the over-predictions were examined, but they were of no particular concern because they were health-protective.

Because monitoring demonstrates what is rather than what might be, a more accurate dose prediction will most likely be obtained if the air concentration that was observed rather than the one that was predicted is used as input to a dose model. The decision to test the assumptions behind the dispersion calculations at the air tritium monitor at the Discovery Center was based on the desire to confirm that air concentrations could be predicted with a degree of accuracy that would demonstrate the validity of the assumptions about release rates and dilution factors. Having demonstrated this in Part 3 of the TDR, the modeling of dose in Part 4 (1953 - 1972) was approached with reasonable confidence.

The doses calculated from observed air concentrations are more accurate and are treated as the true dose in this TDR when they apply to the SW-MEI (i.e., when the SW-MEI was at VIS). When the SW-MEI was at Location T or Q, doses had to be calculated from dispersion modeling alone. Without being able to test the dispersion model at these locations, it is not possible to say whether the resulting doses would be higher (or lower) than they would have been had observed air concentrations been available to use in the dose calculations. Most likely, the magnitude of the uncertainty on the predictions would overlap significantly with the uncertainty on potential observations.

The doses to infant, child, and adult from routine releases that could not have been exceeded for each SW-MEI are summarized in Table 7. Doses for all years are summarized in the Appendix.

Table 7. Highest annual doses (μSv) that could not have been exceeded from routine releases to infant, child, and adult at each location of the SW-MEI, 1953 – 2005.

Location	Year	Infant	Child	Adult
T	1957	590	440	370
VIS (from dispersion modeling)	1964	120	84	71
VIS (from observed HTO in air)	1973	24	16	13
Q	1977	60	42	36

These predicted doses could have been reduced by a factor of two to three had realistic, but still conservative assumptions, about the fraction of the diet that could have been contaminated been used in DCART. Given that the doses based on the assumption of a completely contaminated diet and 100% occupancy were already well below appropriate regulatory dose limits, calculations to reduce them were felt to be unnecessary.

Naturally occurring cosmic and terrestrial radiation at the Livermore site contributes about 600 μSv (60 mrem) annual dose to the public (see the discussions of external radiation measurements in any SAER⁵⁹). Even in 1957, the year of the highest estimated doses, the dose to an infant that could not have been exceeded was at most equal to this naturally occurring exposure that the infant would have received had the Laboratory not existed. For all other years, the dose that could not have been exceeded was a small fraction of the dose from cosmic and terrestrial radiation.

The uncertainty about the mean dose predicted at Location VIS from measured concentrations of HTO in air was primarily due to uncertainty about the measured air concentrations at VIS. When doses were calculated from release rates and dilution factors (i.e., were based on dispersion modeling), the contributors to the uncertainty varied depending upon the circumstances prevailing the year of the release. For example, when releases were from Building 231 and there was high uncertainty about the stack parameters, the dilution factor contributed significantly to the uncertainty. Similarly, the assumptions made about speciation during the early years contributed to the uncertainty. Release rates from specific sources may or may not have significantly affected the uncertainty. The variability on the HTO dose coefficient was important to the uncertainty during the later years of the TDR when dilution factors and release rates became more certain. Predictions were sensitive to the uncertainty about the quantity of milk consumed by an infant, and fruit/fruit vegetable intake and leafy vegetable intake were sometimes important contributors to uncertainty. The uncertainty in the dose predictions averaged 5.6 (upper confidence limit divided by lower confidence limit) for 1973 – 2005 whether or not the doses were predicted from observed or predicted air concentrations, but the uncertainty in the earliest years was as much as a factor of 51 (in 1953).

Doses calculated for this TDR included the contribution from SNL/CA. Releases from SNL/CA were included so that the doses predicted from dispersion modeling could be

⁵⁹ On average in the United States, exposure to naturally occurring radon contributes an additional 2 mSv (200 mrem).

compared with doses predicted from observed concentrations of tritium in air at VIS (which included the contribution of SNL/CA to the atmospheric tritium sampled). The predicted dose drops by about 30% from 1992 through 1994 if the SNL/CA contribution is removed; on average, SNL/CA contributed 13% of the dose at the Discovery Center between 1979 and 1995.

The Laboratory's assessment of dose has generally been reasonable and conservative. The annual doses reported by LLNL never included dose from SNL/CA, and for many years, only inhalation dose was reported. Yet most LLNL dose predictions fell within the confidence intervals of the doses predicted by DCART from release rates and dilution factors. In general it seems that, no matter what doses are accounted for (inhalation, ingestion of HTO, ingestion of OBT), essentially all tritium doses from routine releases fall within a factor of five of each other, even when dispersion models are different.

Diffuse area sources are relatively more important to dose at the perimeter than stack sources because of how the tritium is dispersed by winds close to the ground. The assumptions about area sources, their existence, locations and quantities of tritium released, can have a very large impact on doses. For conservatism, it was important to assume the existence of otherwise unknown area sources, because to neglect to model dose from such an area source might easily result in an underestimation of dose. SNL/CA must have had area sources similar to LLNL's although it is likely the annual release rate(s) from the source(s) would have been much smaller. Certainly, any contribution to dose at the Discovery Center from an SNL/CA area source has been accounted for by using the measured air tritium concentrations at VIS to estimate dose.

DCART estimates concentrations of HTO from HT releases to the atmosphere using empirical factors for the conversion of HT to HTO in the environment that were obtained from experiments carried out in a very different climate from that of Livermore. Nevertheless, it appears that overestimations of HTO in air moisture that might have been expected for years of high HT releases had the empirical conversion factor been too large did not occur. Thus the conversion factors can be applied appropriately to conditions at LLNL.

Doses from accidental releases

The approach for modeling dose from accidental releases in this TDR is unique because the dose model is simply an equation with distributed transfer parameter values based on the performance of the most respected time-dependent tritium model currently in use and on the expert judgment of Wolfgang Raskob, its developer. This approach is the best available under the circumstances given that, except for the most recent accidental releases, there were insufficient meteorological data to run UFOTRI itself.

Because by far the largest potential contributor to ingestion dose after an acute release of tritium is grain and no record of grain growing within 2 or 3 km of LLNL or SNL/CA at the time of the releases has been found (nor is direct personal consumption of locally grown grain likely), the most conservative distribution of doses from each release was calculated on the assumption that the entire diet was contaminated except for grain. A set of assumptions about the quantity of food that could have realistically been contaminated

by the release (that were nonetheless still conservative) was used to determine another confidence interval within which the true dose most likely lay.

When meteorological conditions at the time of the accidental release were known with confidence, the dose that could not have been exceeded was considered to be the 97.5% confidence limit of the dose based on realistic ingestion assumptions averaged with the mean of the dose predictions based on maximum ingestion assumptions (a value very close to the UCL of the realistic dose distribution); when meteorological conditions at the time of the accidental release were unknown, the dose that could not have been exceeded was considered to be the 97.5% confidence limit (i.e., the UCL) when the entire diet was assumed equally contaminated except for grain. The additional conservatism gained from having the entire diet contaminated (except for grain) was used to ensure that the unknowns about the dispersion were probably taken into account. The doses that could not have been exceeded are shown in Table 8. Doses are summarized in more detail in the Appendix.

Table 8. Doses that could not have been exceeded (μSv) to infant, or child, and adult at the locations of the MEI for the years of each accidental release. Doses are ranked from high to low.

Year	Location	Infant/child	Adult
1954 (LLNL)	R	3,000	2,000
1970 (LLNL)	V	170	120
1966 (LLNL)	Q	120	84
1964 (LLNL)	Q	99	64
1965 (LLNL)	X	64	43
1984 (LLNL)	A	11	7.2
1986 (SNL/CA)	L	3.8	2.4
1985 (LLNL)	S	1.6	1.1
1987 (SNL/CA)	K	0.55	0.37

Dose to a child or infant is normally higher than dose to an adult for the same exposure pathways⁶⁰. Dose to a child or infant is estimated to be 40 – 50% higher than dose to an adult for each accidental release examined in this TDR.

Although ingestion dose is expected to be more important than inhalation dose after an accidental release of tritium (particularly HT), most accident consequence models only calculate inhalation dose, because foods have the potential to be monitored before they are eaten. After the 1970 release, extensive vegetation sampling provided a means whereby a maximum and highly unlikely ingestion dose to a child could be estimated. A comparison between doses to adult and child estimated at the time from the 1970 release, doses estimated by ATSDR as part of a PHA of the Livermore site (ATSDR 2004), and doses predicted in this TDR was made. Total doses were very similar, although the relative importance of inhalation and ingestion to the total were different.

⁶⁰ Inhalation dose to an infant is usually lower than that of an adult.

Doses compared with regulatory limits

Recommendations for exposure limits for the public were being developed in the mid- to late-1950's by ICRP and NCRP. On the basis of the 1949 ICRP recommendations on maximum permissible dose to workers, the first recommended limit for an individual member of the public was an annual dose of 1.5 rem (15 mSv) (Jones 2005). In 1957, the NCRP recommended limit to "persons outside of controlled areas" was 500 mrem (5 mSv) (NCRP 1958). This limit was reiterated in 1960 (FRC 1960) and again in 1971 (NCRP 1971), although in both documents there was also mentioned a dose of 170 mrem (1.7 mSv) that applied to an individual in the general population. In 1986 (Office of the Federal Register), the 25 mrem (250 μ Sv) dose limit was introduced for NESHAPs compliance for releases to the atmosphere, and on December 19, 1989, the limit was reduced to 10 mrem. (100 μ Sv) (US EPA 1989).

For the annual doses from routine releases calculated in this TDR, all UCLs on the dose predictions were well below the regulatory requirements of the time. The dose to an infant that was the largest fraction of the regulatory dose limit was 11.8% in 1957 (for a dose of 590 μ Sv and a regulatory limit of 5 mSv); the lowest fraction of dose to an infant compared with the concurrent regulatory limit was 0.18% in 1961 (for a dose of 9.0 μ Sv and a regulatory limit of 5 mSv). Dose to an infant or child, of course, is always greater than to an adult, and the regulations were based on dose to an adult.

A dose from an accidental release should be kept as low as possible, but there is no way to define a limit for such a release. AEC regulations (AEC 1962) required (among other things) reporting releases immediately if members of the public offsite might have received an exposure greater than that set forth in the Radiation Protection Guide for Federal Agencies approved by the President September 20, 1961. These standards only address dose to the thyroid, bone marrow, and bone because they were addressed specifically to ^{131}I , ^{89}Sr , and ^{90}Sr . The smallest of the allowable doses listed was 0.5 rem (5 mSv).

The predicted doses for all accidental releases fell well below this reporting limit that dates to 1962. Even the dose to a child or infant that could not have been exceeded after the 1954 release (about 3 mSv [300 mrem]) was just 60% of the 1962 reporting limit.

The doses predicted in this TDR can also be compared with what is known about the risks from exposure to tritium. Adverse health effects have only been conclusively demonstrated for exposures greater than 10 rem (0.1 Sv) (ATSDR 1999). Numerous studies have also demonstrated that no adverse health effects have been documented for doses less than 360⁶¹ mrem (3.6 mSv) per year (ATSDR 1999). Consequently, ATSDR has defined 360 mrem as a "No Observed Adverse Effect Level". All doses, both from routine and accidental releases fell below this number, including the highest dose that could not have been exceeded, that to a child or infant after the 1954 accidental release. All doses, both from routine and accidental releases, with the exception of the 1954 accidental release and dose to an infant in 1964, fell below what ATSDR has defined as the Minimum Risk Level (MRL) of 100 mrem (1 mSv) that covers acute exposures less than 14 days and chronic exposures of more than a year. 1 mSv (averaged over a five-

⁶¹ Note that the dose from natural background radiation in the Livermore Valley is about 300 mrem (3 mSv) per year.

year period) is also the ICRP (ICRP 1991), NCRP (NCRP 1993), and DOE (DOE Order 5400.5, 1993) dose limit for exposure through all pathways from radiological operations over and above the dose received from natural background radiation by a member of the public. MRLs are intended only to serve as a screening tool to help public health officials decide which release situations require more extensive evaluation. The MRL for chronic exposure to ionizing radiation is considered protective for both cancer and non-cancer health effects.

For the 1954 release, it is likely that the dose received by an actual infant would have been less than the MRL (see Figure 5), given the probability that the plume did not blow over the nearest residence. Even had the dose to an infant been as great as what is called in this TDR “the dose that could not have been exceeded” (i.e., the predicted UCL with maximum ingestion assumptions assumed), the dose would have been no greater than about 3 mSv (300 mrem). In the PHA of the Livermore site (ATSDR 2004), it stated, “Acute exposures to plutonium and tritium via the inhalation, ingestion, and dermal pathways described in this health assessment resulted in cumulative doses of less than 400 mrem [4 mSv] or in chronic exposures less than 100 mrem/year [1 mSv/y] (above background and averaged over five years). These doses are unlikely to produce any adverse health effects and therefore are below levels of public health hazard”. The conclusion from the TDR, therefore, echoes and reinforces, ATSDR’s conclusion that, although some public exposure to tritium releases from LLNL operations probably did occur, estimated maximum exposures were below levels of public health concern, and no adverse health effects would be expected.

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APPENDIX

As has been seen in this summary report, there are many ways to interpret the predicted doses. Although the emphasis in the report was on the upper limit of the dose⁶² (i.e., the dose that could not have been exceeded because of the deliberate choice of conservative assumptions), it is very important to remember that the actual doses to the SW-MEI or the MEI could have been much smaller. Table A1 lists the doses that are considered those that could not have been exceeded from routine and accidental releases for each year. Table A2 lists the mean doses to the SW-MEI and MEI. Table A3 lists the lower limit of possible doses to the SW-MEI or the MEI assuming that a real person actually lived at those locations. The dose to the actual SW-MEI or MEI would probably have been less because it is very likely that the exposed person lived elsewhere and ate less homegrown food. Annual doses to the adult in these tables are those in Figure 3.

Table A1. Doses (μSv) that could not have been exceeded at assumed locations of the SW-MEI and MEI for years (1953 – 2005) of LLNL operations. Doses to the SW-MEI were predicted from release rates using dispersion modeling except for the years 1973 and 1979 – 2005 when doses from routine releases were predicted using mean annual observed concentrations of HTO in air.

Year	Location	Routine releases			Location	Accidental releases	
		Infant	Child	Adult		Infant/child	Adult
1953	T	310	230	190	R	3000	2000
1954	T	310	240	200			
1955	T	320	240	200			
1956	T	230	170	140			
1957	T	590	440	370			
1958	T	320	240	200	Q	99	64
1959	VIS	23	17	14			
1960	VIS	25	18	15			
1961	Q	9.0	6.3	5.4			
1962	VIS	35	25	21			
1963	VIS	49	34	29			
1964	VIS	120	84	71			
1965	VIS	19	13	11			
1966	VIS	25	18	15			
1967	VIS	51	35	30			
1968	VIS	53	36	31	V	120	84
1969	VIS	78	53	45			
1970	VIS	37	26	22			
1971	VIS	23	15	13			
1972	VIS	11	7.6	6.5			
1973	VIS	24	16	13			
1974	Q	19	13	11			
1975	Q	34	24	21			
1976	Q	38	25	22			
1977	Q	60	42	36			
1978	Q	40	28	24			

⁶² This summary report has demonstrated that no adverse health effects from LLNL operations are expected even when it is assumed that the SW-MEI or ME received the highest possible dose.

Table A1 continued

Year	Location	Routine releases			Accidental releases		
		Infant	Child	Adult	Location	Infant/child	Adult
1979	VIS	15	10	8.6			
1980	VIS	15	10	8.9			
1981	VIS	8.8	6.0	5.1			
1982	VIS	14	9.1	7.9			
1983	VIS	11	7.1	6.1			
1984	VIS	13	8.4	7.3	A	11	7.2
1985	VIS	9.1	6.2	5.3	S	1.6	1.1
1986	VIS	10	7.1	6.0			
1987	VIS	19	13	11			
1988	VIS	15	10	8.9			
1989	VIS	11	7.2	6.2			
1990	VIS	6.2	4.2	3.6			
1991	VIS	4.2	2.9	2.5			
1992	VIS	1.5	1.0	0.84			
1993	VIS	1.4	0.93	0.79			
1994	VIS	0.85	0.57	0.49			
1995	VIS	0.78	0.54	0.46			
1996	VIS	1.6	1.1	0.97			
1997	VIS	2.1	1.4	1.2			
1998	VIS	0.74	0.51	0.43			
1999	VIS	1.3	0.85	0.74			
2000	VIS	0.48	0.32	0.28			
2001	VIS	0.28	0.18	0.16			
2002	VIS	0.35	0.24	0.21			
2003	VIS	1.0	0.65	0.56			
2004	VIS	0.24	0.16	0.13			
2005	VIS	0.30	0.21	0.18			

Table A2. Mean doses (μSv) potentially received at assumed locations of the SW-MEI and MEI for years (1953 – 2005) of LLNL operations. Doses to the SW-MEI were predicted from release rates using dispersion modeling except for the years 1973 and 1979 – 2005 when doses from routine releases were predicted using mean annual observed concentrations of HTO in air. Doses to the MEI were based on conservative assumptions when meteorology was unknown (1954, 1964, 1966, and 1984) and on realistic assumptions when it was known (1965, 1970, 1985). The mean doses based on conservative assumptions are not shown in Figure 5.

Year	Location	Routine releases			Accidental releases		
		Infant	Child	Adult	Location	Infant/child	Adult
1953	T	24	20	17			
1954	T	26	22	18	R	480	330
1955	T	30	27	22			
1956	T	26	23	19			
1957	T	48	42	34			
1958	T	36	31	26			
1959	VIS	2.7	2.6	1.9			
1960	VIS	2.3	2.0	1.6			

Table A2 continued

Year	Location	Routine releases			Accidental releases		
		Infant	Child	Adult	Location	Infant/child	Adult
1961	Q	3.2	2.8	2.3			
1962	VIS	13	11	8.8			
1963	VIS	18	15	1.2			
1964	VIS	40	35	29	Q	18	12
1965	VIS	6.7	5.8	4.7	X	8.7	6.1
1966	VIS	8.9	7.7	6.3	Q	19	13
1967	VIS	18	16	13			
1968	VIS	19	17	13			
1969	VIS	27	24	19			
1970	VIS	13	11	9.2	V	23	17
1971	VIS	8.0	7.0	5.7			
1972	VIS	4.1	3.6	2.9			
1973	VIS	8.5	7.3	6.0			
1974	Q	6.7	5.7	4.7			
1975	Q	12	11	8.6			
1976	Q	13	12	9.5			
1977	Q	21	18	15			
1978	Q	14	13	10			
1979	VIS	5.7	4.9	4.0			
1980	VIS	5.8	5.1	4.1			
1981	VIS	3.3	2.9	2.4			
1982	VIS	2.2	4.5	3.7			
1983	VIS	4.0	3.5	2.8			
1984	VIS	4.8	4.2	3.4	A	2.2	1.5
1985	VIS	3.5	3.0	2.4	S	0.22	0.16
1986	VIS	3.8	3.3	2.7			
1987	VIS	7.1	6.2	5.0			
1988	VIS	5.6	4.8	4.0			
1989	VIS	4.0	3.5	2.8			
1990	VIS	2.4	2.0	1.7			
1991	VIS	1.6	1.4	1.1			
1992	VIS	0.55	0.48	0.39			
1993	VIS	0.52	0.45	0.37			
1994	VIS	0.32	0.28	0.23			
1995	VIS	0.30	0.26	0.21			
1996	VIS	0.64	0.55	0.45			
1997	VIS	0.79	0.69	0.56			
1998	VIS	0.28	0.24	0.20			
1999	VIS	0.49	0.42	0.34			
2000	VIS	0.18	0.16	0.13			
2001	VIS	0.11	0.091	0.074			
2002	VIS	0.13	0.11	0.093			
2003	VIS	0.39	0.34	0.28			
2004	VIS	0.088	0.076	0.062			
2005	VIS	0.11	0.093	0.076			

Table A3. Doses (μSv) representing the lower limit of those that could have been received at assumed locations of the SW-MEI and MEI for years (1953 – 2005) of LLNL operations. Doses to the SW-MEI were predicted from release rates using dispersion modeling except for the years 1973 and 1979 – 2005 when doses from routine releases were predicted using mean annual observed concentrations of HTO in air. The lower limit for accidental releases is the 2.5% confidence limit based on realistic ingestion assumptions.

Year	Location	Routine releases			Location	Accidental releases	
		Infant	Child	Adult		Infant/child	Adult
1953	T	4.2	4.7	3.8	R	1.6	1.8
1954	T	4.6	5.1	4.1			
1955	T	5.6	6.1	4.9			
1956	T	5.2	5.8	4.8			
1957	T	13	15	12			
1958	T	8.1	9.4	7.6	Q	0.092	0.090
1959	VIS	0.67	0.77	0.62			
1960	VIS	0.52	0.60	0.49			
1961	Q	0.83	0.98	0.80			
1962	VIS	3.1	3.5	2.8			
1963	VIS	4.3	5.0	4.0			
1964	VIS	8.4	9.2	7.4			
1965	VIS	1.7	1.9	1.6			
1966	VIS	2.0	2.3	1.9			
1967	VIS	4.7	5.4	4.4			
1968	VIS	4.9	5.9	4.7	V	0.14	0.13
1969	VIS	6.7	7.8	6.3			
1970	VIS	2.9	3.4	2.7			
1971	VIS	2.0	2.3	1.9			
1972	VIS	1.2	1.4	1.1			
1973	VIS	2.0	2.3	1.9	Q	0.026	0.022
1974	Q	1.7	2.0	1.6			
1975	Q	3.1	3.6	2.9			
1976	Q	3.6	4.3	3.4			
1977	Q	5.5	6.3	5.2			
1978	Q	4.0	4.6	3.8			
1979	VIS	1.7	2.0	1.6			
1980	VIS	1.7	2.0	1.7			
1981	VIS	1.0	1.2	0.94			
1982	VIS	1.5	1.9	1.5			
1983	VIS	1.1	1.3	1.1	A	0.0076	0.0079
1984	VIS	1.4	1.7	1.4			
1985	VIS	1.0	1.2	0.95			
1986	VIS	0.90	1.0	0.83			
1987	VIS	2.1	2.5	2.1			
1988	VIS	1.5	1.8	1.5	S	0.012	0.0076
1989	VIS	1.2	1.3	1.1			
1990	VIS	0.70	0.83	0.67			
1991	VIS	0.48	0.57	0.47			
1992	VIS	0.16	0.19	0.16			
1993	VIS	0.15	0.19	0.15			

Table A3 continued

		Routine releases			Accidental releases		
Year	Location	Infant	Child	Adult	Location	Infant/child	Adult
1994	VIS	0.093	0.11	0.089			
1995	VIS	0.085	0.10	0.082			
1996	VIS	0.19	0.22	0.18			
1997	VIS	0.24	0.28	0.23			
1998	VIS	0.075	0.087	0.070			
1999	VIS	0.15	0.18	0.15			
2000	VIS	0.051	0.061	0.050			
2001	VIS	0.031	0.038	0.030			
2002	VIS	0.035	0.040	0.033			
2003	VIS	0.13	0.16	0.13			
2004	VIS	0.025	0.029	0.024			
2005	VIS	0.023	0.026	0.021			

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ABBREVIATIONS, ACRONYMS AND DEFINITIONS

AEC	Atomic Energy Commission
AIRDOS-EPA	The first of the EPA's CAACs (Moore et al. 1979)
ATSDR	Agency for Toxic Substances and Disease Registry
BIOMASS	B iospheric M odel A ssessment, a coordinated research program of the IAEA
CAAC	The EPA's Clean Air Act Code
CAP88-PC	The CAAC code used by LLNL for compliance with NESHAPs (Parks 1992)
CPS	Continuous Point Source (code) (Peterson 1976)
DCART	D oses from C hronic A tmospheric R eleases of T ritium; a steady-state, stochastic dose model (Peterson 2006)
Dilution Factor	A term that refers to the air concentration for unit source strength (or χ/Q); units are $\text{Bq m}^{-3} / \text{Bq s}^{-1}$ (or $\text{Ci m}^{-3} / \text{Ci s}^{-1}$). The term, although standard for χ/Q , can be misleading, because the higher the dilution factor, the higher the air concentration.
Distribution	A function of a discrete random variable yielding the probability that the variable will have a given value. Types of distributions include
	Lognormal The probability distribution of any random variable whose logarithm is normally distributed. It can be expressed as a geometric mean and geometric standard deviation. It is commonly used for dilution factors.
	Normal A theoretical frequency distribution for a set of variable data, usually represented by a bell-shaped curve symmetrical about the mean and is expressed as a mean and standard deviation. Also called <i>Gaussian distribution</i> . Source terms are commonly distributed normally.
	Triangular A distribution with three terms: minimum, likely, and maximum.
	Uniform A distribution in which all values in the range have an equal probability of being sampled
DOE	Department of Energy

ABBREVIATIONS, ACRONYMS AND DEFINITIONS *continued*

DT	d euterium h ydrogen gas
DTO	a form of water in which the hydrogen is replaced by deuterium and tritium
DWTF	D econtamination and W aste T reatment F acility; also an air tritium sampler
EMRAS	E nvironmental M odeling for R adiation S afety, a coordinated research program of the IAEA
EPA	E nvironmental P rotection A gency
ERDA	E nergy, R esearch and D evelopment A ministration
Evap	Evaporation (trays)
FRC	F ederal R adiation C ouncil
HOTSPOT	Accident consequence model (Homann 1994)
HT	Tritiated hydrogen gas
HTO	Tritiated water
IAEA	I nternational A tomc E nergy A gency
ICRP	I nternational C ommission on R adiation P rotection
Incin	Incinerator
ITER	international experimental fusion reactor
LLNL	L awrence L ivermore N ational L aboratory
MEI	M aximally E xposed I ndividual (to releases from one facility)
MRL	M inimum R isk L evel (1 mSv [100 mrem]) as defined by ATSDR)
NCRP	N ational C ommittee on R adiation P rotection and Measurements
NESHAPs	N ational E mission S tandards for H azardous A ir P ollutants (40 CFR 61 Subpart H. (National Emission Standards for Emissions of Radionuclides other than Radon from Department of Energy Facilities).
OBT	o rganically b ound t ritium
PHA	P ublic H ealth A ssessment
P/O	P redicted-to- o bserved (ratio)
SAER	LLNL' Site A nnual E nvironmental R eport
SNL-CA	S andia N ational L aboratories, Livermore, CA

ABBREVIATIONS, ACRONYMS AND DEFINITIONS continued

SW-MEI	S ite- w ide m aximally e xposed i ndividual (at the off-site location where the air concentration from all sources of tritium is the greatest)
TDR	T ritium D ose R econstruction (LLNL, 1953 – 1972, both routine and accidental releases)
TFWT	t issue f ree w ater t ritium, or the water in a plant
TRL	T ritium R esearch L aboratory (SNL/CA)
UCL	u pper confidence limit of a 95% (normally) distribution of dose predictions (i.e., the 97.5% confidence limit)
UFOTRI	U nfall f olgenmodell für T ritium f reisetzungen; a program for assessing the off-site consequences from accidental releases (Raskob 1990, 1993)
UNCLE	U Niversity of California L awrence Livermore Laboratory E mployees C redit Union
USGS	U nited States G eological Survey
VIS	Location of an air tritium sampler near the Discovery Center
WAA	W aste A ccumulation A rea